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RECONSTRUCTION OF ^{131}I AND ^{137}Cs RADIOACTIVE CONTAMINATION IN UKRAINE CAUSED BY THE CHERNOBYL ACCIDENT USING ATMOSPHERIC TRANSPORT MODELING

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Abstract. The work is devoted to the reconstruction of time-dependent ^{137}Cs and ^{131}I contamination fields in the territory of Ukraine in the initial period of the Chernobyl accident using the model of atmospheric transport LEDI (Lagrangian-Eulerian DIffusion model). The radioactivity atmospheric transport over the territory of Ukraine was simulated during the first 12 days after the accident (from 26 April to 7 May 1986) using real aerological information and rain measurement network data. The detailed scenario of the release from the accidental unit of the Chernobyl nuclear plant has been built (including time-dependent radioactivity release intensity and time-varied height of the release). The modeling results were compared with available ^{137}Cs air and ground contamination measurement data, just as with available data of radioiodine daily deposition measurements made at the network of meteorological stations in Ukraine and data of the assessments of ^{131}I soil contamination obtained from the ^{129}I measurements. The conducted calculations have enabled to explain the main features of spatial and temporal variations of radioactive contamination fields over the territory of Ukraine on the regional scale, including the formation of the major large-scale spots of radioactive contamination caused by dry and wet deposition. The airborne ^{131}I concentration and ground deposition fields were calculated as the database for subsequent thyroid dose reconstruction for inhabitants of radioactive contaminated regions.

Key word index: accidental release, dry deposition, wet deposition, radionuclides, atmospheric transport.

1. INTRODUCTION

The way to solve the problem of the reconstruction of both the total iodine deposition in the territories contaminated after the Chernobyl accident and time variability of activity in air and soil is depend on the available information. The data of direct measurements of ^{131}I soil contamination made in Belarus in 1986 give reasons to construct the map of ^{131}I deposition of the territory of this country (Germenchuk *et al*., 1996). Unfortunately, very few direct measurements of ^{131}I contamination in soil have been made in 1986 in the territory of Ukraine. So there are some approaches to reconstruct the total iodine isotopes activity in soil. The most simple method is the assumption about the correlation between ^{131}I and ^{137}Cs contamination. The obtained empirical relations between iodine and cesium deposition values were used for the reconstruction of thyroid doses in Ukraine (Likhtarev *et al*., 1996) and in Russia (Makhon'ko *et al*., 1996). But due to the large variability of ^{131}I and ^{137}Cs ratio in the Chernobyl's deposition this approach can result in large uncertainties of the reconstructed iodine contamination. The another way is the estimation of ^{131}I deposition densities using the direct measurements of long-lived ^{129}I soil contamination, known isotopic ratio for these isotopes and estimated pre-Chernobyl background of ^{129}I (Michel R. *et al*., 2004). Makhon'ko *et al*. (1996) used the measurements of ^{131}I daily deposition on collectors exposed at network of meteorological stations in April-May 1986 for the reconstruction of iodine

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isotopes deposition in the territory of the former USSR. Compared with previous methods this one gives possibility to reconstruct not only cumulative iodine deposition but the daily dynamics of air and soil contamination also. But in the territory of three most contaminated regions of Ukraine (Kyiv, Zhytomyr, Chernigiv ones) there are only two meteorological stations where the ^{131}I deposition have been measured (see later).

So the method of modeling of the Chernobyl radioactivity release atmospheric transport can be useful instrument for the reconstruction of the dynamics of ^{131}I fallout on the territory of Ukraine. It is based on the use of mesoscale atmospheric transport model LEDI (Buikov *et al*., 1992; Talerko and Kuzmenko, 1999). The first step of this work was the verification of used approach for the task of modeling of ^{137}Cs transport and deposition in Ukraine (chapter 2). It enables to specify the volatile radionuclides release scenario from the accidental unit of the Chernobyl nuclear power station (taking into account the time variability of its parameters) and to assess the large and small scale features of radioactivity deposition field in the territory of Ukraine on the base of the comparison of obtained results with available data of soil cesium contamination measurements.

The main aim of this work was the calculation of ^{131}I air and surface contamination dynamics in the territory of Ukraine for the purpose of the thyroid dose reconstruction carried out as a part of joint Ukrainian-American project “UkrAm Study of Thyroid Cancer and Other Thyroid Diseases in Ukraine Following the Chornobyl Accident” sponsored with National Cancer Institute now. Modern retrospection in Ukraine takes into account the assessment of territories contamination dynamics into the system of retrospective dosimetry of thyroid exposure. In the initial period of the accident the direct thyroid activity measurements have been made for the some part of the population lived in the most contaminated Kyiv, Zhytomyr and Chernigiv regions only. The data of the thyroid dose reconstruction for the rest of peoples of these regions and another territory of Ukraine may be obtained now on the base of the atmospheric transport modeling only. The results of the activity transport simulation enable to use some ecological models for the thyroid dose estimation taking into account that the dose estimations are considerably dependent on time-variable air-borne concentration values, deposition values and especially on the activity arrival time and the finishing of fallout.

Main tasks of ^{131}I transport modeling for the territory of Ukraine were:

- To reconstruct the formation features of time-dependent airborne and deposited ^{131}I fields in the territory of Ukraine during the initial stage of the Chernobyl accident (26 April - 7 May 1986);

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- To explain mechanisms of formation of large-scale spots of radioactivity deposition in Ukraine (including relative contribution of dry and wet deposition for separate spots);
- To create the database of airborne and deposit ^{131}I concentration values for the inhabitant places in contaminated territories of Ukraine for the purposes of thyroid dose reconstruction using obtained modeling results;
- To create the background for the assessment of the contribution of short-lived iodine isotopes using the atmospheric transport modeling.

2. MESOSCALE MODELING OF ^{137}CS CONTAMINATION FORMATION IN UKRAINE CAUSED BY THE CHERNOBYL ACCIDENT

2.1. Problem of mathematical modeling of the Chernobyl release atmospheric transport

After the Chernobyl accident on 26 April 1986, a lot of works on numerical modeling of transport of a radioactive release from the 4-th unit of the Chernobyl nuclear power plant was published. They are devoted to the assessment of an air and underlying surface contamination following the accidental release. The interest to such simulations is of two kinds. On the one hand, the scales of the catastrophe have stipulated a necessity of an reliable evaluation of an air, ground, vegetation radioactive contamination for distances in a wide interval from hundreds meters up to thousands kilometers from the source, while the available measurement data don't ensure satisfactory description of a radioactive contamination formation neither in space, nor in time. The method of mathematical modeling in this situation becomes one of main ways of the accident course reconstruction and evaluation of its consequences for the environment. On the other hand, the measurement data of radioactive contamination after the accident are a unique material for verification of the atmospheric transport models, in particular regional and long-distance ones (project ATMES) (Klug *et al*., 1992).

The problem of reconstruction of the time dependence of near-ground air concentration of main dose-forming radionuclides in the initial period of the Chernobyl accident, as well as the dynamics of the radioactive fallout, remains an important and still unsettled question. The methods of mathematical modeling of atmospheric transport of radioactive release and its deposition on the underlying surface are an important tool for the support of retrospective dosimetry studies. Such approach should be based on the use of the following components:

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- mathematical model of atmospheric transport that enables to describe a transport of the radioactive release from the Chernobyl NPP on a mesoscale and regional scale;
- meteorological data during the initial period of the accident;
- unavoidable additional hypotheses and assumptions concerning the parameters of the source term for the Chernobyl release (first of all, the dynamics of the radioactive release in the first 10 days following the accident).

The Chernobyl catastrophe is an example of an accident involving a high temperature release of large amounts of radioactivity and resulting in radioactive contamination of the territories on distances of thousands kilometers from the source. In this case it is necessary to consider the problem of modeling of atmospheric transport of the Chernobyl release separately for several different space scales, namely: 1) long-distance transport of a radioactivity (see, e.g. Smith and Clark, 1988; Haas *et al.*, 1990; ApSimon and Wilson, 1989; Albergel *et al.*, 1988; Gudiksen *et al.*, 1989), 2) radioactivity transport on a regional scale (Izrael *et al*., 1989, Izrael *et al*., 1990, Pitkevich *et al.*, 1994), 3) pollutant dispersion within near-source zone (the space scale is about tens kilometers) (Izrael *et al*., 1987; Vakulovsky *et al.*,1993). For each of them it is necessary to use different mathematical models, specific to each scale, and various sets of an input information about meteorological conditions and release parameters.

The most complicated task from physical and mathematical points of view is modeling of pollutant transport on a regional scale (on distances up to several hundreds kilometers). It is necessary in this case to take into account essential time-dependence (diurnal variations of the atmospheric boundary layer characteristics or the weather variations) of conditions of pollutant dispersion in an atmosphere, and also space heterogeneity of fields of the meteorological elements. On the other hand, the reasonable compromise should be found in a model between both a degree of the account of basic physical processes which defines pollutant dispersion, the choice of suitable computing algorithm which is used to solve the equations of pollutant transport and possibilities of used computers.

Taking this into consideration, the regional model of atmospheric pollutant transport LEDI (Lagrangian-Eulerian DIffusion model) (Buikov *et al*., 1992, Talerko and Kuzmenko, 1999) is used for the reconstruction of dynamics of radioactive contamination of Ukraine during an initial phase of the Chernobyl accident. As applied to the description of the formation of initial radioactive contamination fields in the territory of Ukraine on the regional scale is to account for the formation mechanisms of large-scale radioactive deposition spots, in particular well-known ^{137}Cs spots of quite large (over 37 kBq m^{-2}) deposition density in central (Kyiv, Zhytomyr, Cherkasy), western (Rivne, Lutsk) and southern (Cherkasy, Vinnytsia, Khmel'nyts'kyi) regions of Ukraine. The

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obtained results can be used as the basis for subsequent reconstruction of space and time variations of short-lived volatile radionuclides contamination especially iodine isotopes.

2.2. Brief model description

The LEDI model has been developed for the calculations of pollutant transport at distances up to 1000 km from a gaseous or aerosol point source with the effective release height varied from 0 to 1500 m. It takes into account spatial inhomogeneity and time-dependence of pollutant transport conditions, different types of a pollutant source (stationary, time-dependent or instantaneous), physical and chemical release composition (gas, aerosol particles), isotope composition, horizontal heterogeneity of an underlying surface (moderately complex terrain and inhomogeneous vegetative cover).

In the model the combination of Lagrangian and Eulerian methods is used to describe a pollutant transport in an atmospheric boundary layer. Such approach allows physically correctly to take into account in the model major factors, defining pollutant transport with rather small expenses of computer time. For time-dependent source the release is modeling as a set of puffs sequential in time. The horizontal puff trajectory is computed using the methods of spherical geometry (Petterssen, 1956) because of Earth surface curvature could be taken into consideration. It is determined by a set of points with geographical coordinates φ_i, ω_i (i= 0, 1, ..., φ_0, ω_0 - source coordinates). The transport velocity of puff number m of isotope number r and particle size range f on the i-th time step U_{mi}^{rf} is defined as mean horizontal wind velocity weighted over pollutant transport layer (as rule, it is all boundary layer $0 < z < B$) with multipliers proportional to the pollutant concentration as a function of the height:

$$\vec{U}_{mi}^{rf}(\varphi_i,\omega_i,t) = \frac{\int_o^B C_m(\varphi_i,\omega_i,z,t)\vec{u}(\varphi_i,\omega_i,z,t)dz}{\int_o^B C_m(\varphi_i,\omega_i,z,t)dz}, \qquad (2.1)$$

where $\vec{u}(\varphi_i,\omega_i,z,t)$ is the wind velocity (m s^{-1}), $C_m(\varphi, \omega, z, t)$ is the pollutant concentration (Bq m^{-3}).

On the each i-th trajectory calculation step the Cartesian coordinate system is introduced, where x, y, z are the coordinates in the direction of the puff transport, in the horizontal direction perpendicular to the transport and in the vertical direction respectively. Then for the fixed point with Cartesian coordinates x, y, z (or corresponding geographical φ, ω, z) the combined Lagrangian

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- Eulerian method allows to calculate the time-dependent volume concentration field $C_m(\varphi, \omega, z, t)$ determined by the transport of m-th puff. In experimental methods of measurements the radioactive particles are accumulated in samplers during some sampling period, i.e. the integral flux over time $J = \int_{t_1}^{t_2} U_m \cdot C_m^{'} \cdot dt$ is measured. For the constant transport velocity U_m it depends on the integral of volume concentration over time $C(\varphi, \omega, z) = \int_{t_1}^{t_2} C_m^{'}(\varphi, \omega, z, t) \cdot dt$ (Bq s m^{-3}). It was calculated from

$$C_m^{rf}(\varphi, \omega, z) = \chi_{mi}^{rf}(z)\Big|_{\substack{\varphi=\varphi_i \\ \omega=\omega_i}} \cdot \frac{\exp\left[-y_i^2 / 2(\sigma_{mi}^{rf})^2\right]}{\sqrt{2\pi} \cdot U_{mi}^{rf} \cdot \sigma_{mi}^{rf}}, \tag{2.2}$$

where $\chi_{mi}^{rf}(z)$ is the vertical profile of the radionuclide one-dimensional concentration (Bq m^{-1}) calculated at a point with coordinates (φ_i, ω_i), φ, ω are the geographical coordinates of the points in the plane perpendicular to transport direction, y_i is the distance (m) between points with coordinates (φ_i, ω_i) and (φ, ω).

For polydisperse pollutant the size distribution is divided into several size ranges and the calculations are performed for each size range separately.

The semiempirical one-dimensional turbulent diffusion equation is used for the calculation of vertical profile of the one-dimensional particle concentration χ_{mi}^{rf} in the m-th puff:

$$\frac{\partial \chi_{mi}^{rf}}{\partial t} = \frac{\partial}{\partial z}\left(k_z \cdot \frac{\partial \chi_{mi}^{rf}}{\partial z}\right) + W_g^f \cdot \frac{\partial \chi_{mi}^{rf}}{\partial z} - \Lambda \cdot \chi_{mi}^{rf} - \lambda \cdot \chi_{mi}^{rf} - w \cdot \frac{\partial \chi_{mi}^{rf}}{\partial z} \tag{2.3}$$

using the boundary conditions

$$k_z \frac{\partial \chi_{mi}^{rf}}{\partial z}\Big|_{z=B} = 0, \tag{2.4}$$

$$k_z \frac{\partial \chi_{mi}^{rf}}{\partial z} + W_g^f \cdot \chi_{mi}^{rf}\Big|_{z=0} = V_s^f \chi_{mi}^{rf} \tag{2.5}$$

and the initial condition

$$\chi_{mo}^{rf}\Big|_{t=0} = Q_m^{rf} \delta(z - h_{ef}), \tag{2.6}$$

where $\delta(z)$ is Dirac delta function, k_z is the vertical eddy diffusion coefficient (m^2 s^{-1}), λ is the half-life time of radioactive decay (s^{-1}), Λ is the washout ratio (s^{-1}), V_s^f is the dry deposition velocity (m

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s^{-1}), Q_m^{rf} is the radionuclide emission amount in the m-th puff (Bq), W_g^f is sedimentation velocity (m s^{-1}) of the particle of a given size range, h_{ef} is the effective source height (m), B is the boundary layer height (m), w is the vertical wind in the boundary layer (m s^{-1}).

The equation (2.3) with conditions (2.4) - (2.6) is solved numerically. The integration is performed using the Runge - Kutta method. The model has some vertical layers of unequal thickness between the ground and the boundary layer height. The values of Dz were chosen from 10 m near the surface to 20 - 100 m in the upper part of boundary layer depending on it's thickness. To describe the diffusion on the boundaries two 'artificial' grid points were introduced.

The sedimentation velocity of aerosol particles is given by

$$W_g^f = \begin{cases} 3\cdot 10^4 \rho\, d^2, & d < 1.15\cdot 10^{-3}\,\rho^{-1/3}; \\ 34\rho^{2/3} d, & 1.15\cdot 10^{-3}\,\rho^{-1/3} \le d \le 2.13\cdot 10^{-2}\,\rho^{-1/3}; \end{cases} \qquad (2.7)$$

where ρ is the particle density (kg m^{-3}), d is the particle diameter (m).

The washout ratio is a function of the rainfall rate J (mm hr^{-1}) as follows:

$$\Lambda = c^r J\,, \qquad (2.8)$$

where c^r =2.6 10^{-5} hr mm^{-1} s^{-1} for aerosol particles.

Following (2.2), the pollutant concentration distribution is assumed to be Gaussian in the direction perpendicular to the puff transport vector $\vec{U}_{mi}^{rf}$. For the distances up to 20 km the cross-wind standard deviation $\left[\sigma_{mi}^{rf}(l)\right]^2$ (where $l = \int_0^T \left|U_{mi}^{rf}\right| \cdot dt$ is the travel distance of puff number m within the period T) is expressed using the standard expression where the constant c_3 depends on the atmospheric Pasquill stability class (Briggs, 1974):

$$\sigma_{mi}^{rf}(l) = \frac{c_3 \cdot l}{\sqrt{1 + 10^{-4}\cdot l}} \qquad \text{for} \quad l \le l_1 = 20 \text{ km}, \qquad (2.9)$$

Following Byzova *et al.* (1991) the lateral plume dispersion is presented for distances over 40 km as a sum of contributions due to horizontal turbulent diffusion (the first term in right-side) and due to lateral dispersion caused by the wind direction shear in the boundary layer (the second one):

$$\sigma_{mi}^{rf}(l) = \left(2\cdot\overline{\sigma_v^2}\cdot\tau_L^2\cdot\left(\frac{l}{U_{mi}^{rf}\cdot\tau_L} - \frac{3}{2} - \frac{1}{2}\cdot\exp\left(-\frac{2l}{U_{mi}^{rf}\cdot\tau_L}\right) + \right.\right.$$

$$\left.\left. + 2\cdot\exp\left(-\frac{l}{U_{mi}^{rf}\cdot\tau_L}\right)\right) + 2.95\cdot 10^{-5}\cdot(\Delta\psi)^2\cdot l^2\right)^{1/2} \quad \text{for} \quad l \ge l_2 = 40\,\text{km}. \qquad (2.10)$$

In the range from 20 to 40 km the lateral plume dispersion is calculated by interpolation.

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The windshear angle in the boundary layer $\Delta\psi$ is calculated following Orlenko (1979):

$$\Delta\psi = \left(67.8 - 6.2\log\left(\frac{U_g}{Fz_0}\right)\right)\cdot\Phi(\mu_0), \tag{2.11}$$

where U_g is the geostrophic wind speed (m s^{-1}), F is the Coriolis parameter (s^{-1}), $\Phi(\mu_0)$ is the dimensionless function of a stability parameter $\mu_0 = g(T_0 - T_B - \gamma B)/(\overline{T}FU_g)$, where g is the acceleration of gravity (m s^{-2}), T_0 is the soil temperature (K), T_B is the temperature at the top of boundary layer (K), $\overline{T}$ is the mean temperature in the boundary layer (K), γ is the temperature gradient in the free atmosphere ($6.5\cdot10^{-3}$ K m^{-1}).

The lateral component of turbulent energy $\overline{\sigma_i^2}$ (m^2 s^{-2}) and the Lagrangian integral time scale of atmospheric turbulence t_L (s) averaged over the boundary layer (BL) in (2.12) are calculated according Byzova *et al.* (1991):

$$\overline{\sigma_v^2} = \begin{bmatrix} 0.157w_*^2 \\ 0.84u_*^2 \\ 1.02u_*^2 \end{bmatrix} \times \left(\frac{t}{T_m}\right)^{0.35} \begin{matrix} , \\ , \\ , \end{matrix} \quad \begin{matrix} \text{for unstable BL,} \\ \text{for neutral BL,} \\ \text{for stable BL;} \end{matrix} \tag{2.12}$$

$$\tau_L = \begin{cases} 0.5z_i/w_*, & \text{for unstable BL,} \\ \overline{\sigma_v^2}/0.6\overline{\varepsilon}, & \text{for neutral and stable BL,} \end{cases} \tag{2.13}$$

where u_* is the friction velocity (m s^{-1}), w_* is the convective velocity (m s^{-1}), z_i is the mixed layer height (m), $\overline{\varepsilon}$ is the dissipation of turbulent kinetic energy (m^2 s^{-3}). The last factor in (2.12) represents the correction coefficient due to the difference between the period of pollutant diffusion t and the period of averaging of used experimental data T_m.

The value of $\overline{\varepsilon}$ is estimated as follows:

$$\overline{\varepsilon} = \begin{cases} \dfrac{0.5w_*^3}{z_i} + \varepsilon(z_i) & \text{for unstable BL,} \\ 2.5\dfrac{u_*^3}{B}\left(\ln\dfrac{B}{z_0} - 1.8\right) + \varepsilon(B) & \text{for neutral BL,} \\ 2.5\dfrac{u_*^3}{B}\left(\ln\dfrac{2B}{z_0} - 3.57\right) + \varepsilon(B) & \text{for stable BL,} \end{cases} \tag{2.14}$$

where z_0 is the roughness height (m), $\varepsilon(z_i)=10^{-4}$ m^2 s^{-3} ,$\varepsilon(B)=4\ 10^{-4}$ m^2 s^{-3}.

The vertical eddy diffusion coefficient k_z in the surface layer was assumed to equal to the eddy diffusion one for momentum (Nieuwstadt and van Dop, 1982) as the function depending on friction velocity u_* and the Monin - Obukhov length L. The height of the surface layer h was calculated following Yordanov (1977). Within the planetary layer the k_z profile was obtained by

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fitting a quadratic polynomial between coefficient values at the top of the surface layer and to the one for free atmosphere at the top of the boundary layer taken equals to zero. The values of u_* and L have been calculated according Orlenko (1979).

Because of the pollutant dispersion takes place under inhomogeneous and time-dependent meteorological conditions the turbulent parameters calculations are repeated in each step of the puff trajectory calculations as describe above.

The deposition density $D_m^{rf}(\varphi,\omega)$ caused by the transport of radionuclide number r in the m - th puff that consists of particles with f - th size range in the point (φ, ω) is defined by

$$D_m^{rf}(\varphi,\omega) = V_s^f \cdot C_m^{rf}(\varphi,\omega,0) + \Lambda \int_0^B C_m^{rf}(\varphi,\omega,z)\cdot dz\,. \qquad (2.15)$$

The former term on the right hand side defines the contribution in the deposition density due to dry deposition of the pollutant on the underlying surface and the latter due to wet deposition by rain from the boundary layer.

The total amounts of the time integral of volume concentration and the ground deposition density in the point (φ, ω) for some period are defined as the sums of the individual contributions from the puffs passed over this point during this period.

It was supposed that the ^{137}Cs contamination of the territory Ukraine outside local zone near the Chernobyl power plant is formed mainly by small aerosol particles. Thus in following ^{137}Cs transport simulations the only one particle size range was used with size d=1 μm and density ρ=2.5 10^3 kg m^{-3}. The dry deposition velocity value of 5 10^{-3} m s^{-1} was taken according measurements data in Ukraine (Israel *et al*., 1990). The set of topography and landuse data was used with horizontal resolution 10 km.

2.3. Meteorological conditions of radioactivity transport

To describe the transport of the radioactive release in the initial period of the Chernobyl accident, the data of radiosounding measurements of the atmosphere performed by a network of aerological stations of the State Committee for Meteorology ("Goskomhidromet") of the former USSR have been used as input meteorological information in the release transport modeling. They included measured vertical profiles of temperature, pressure, wind velocity and direction in a number of radiosounding stations located throughout the territory of Ukraine (9 stations among which Kyiv and Shepetivka being the nearest ones to the Chernobyl NPP and areas of intensive radioactive contamination), as well as those of Gomel (Belarus), and Kursk (Russia). The average

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distance between the stations is about 250 km, what may be considered as a satisfactory value to be used in subsequent modeling.

Sounding was being performed, in most of places, 4 times per day at 0, 6, 12 and 18 h GMT, but in some of them measurements were made according a short program twice at 0 h and 12 h (Kryvy Rig, Uzhgorod, Kharkiv) or 3 times excluding 18 h (Odesa, Chernivtsi) every day. Data about air temperature, wind direction and velocity, atmospheric pressure from the ground surface to an altitude of 3 km have been used for the simulations. The measurements data were used at standard altitudes: 0.1; 0.2; 0.3; 0.5; 0.6; 0.9; 1.0; 1.5; 2.5 km above the ground surface and on isobaric surfaces: 1000, 850, 700 hPa. Besides, the values of the above parameters have been used which corresponded to the position of inversion layers and special wind points.

For stations where radiosounding measurement was not being performed in a certain time interval the missing information was compensated by data obtained from the All-Union Research Institute of Hydrometerological Information - World Data Center) in Obninsk (Russia). In this case the data contained the values of wind velocity on standard isobaric surfaces – 700 hPa and 850 hPa, and at altitudes of 300 m, 600 m, 900 m, 1000 m, and 2000 m. Information on vertical profiles of temperature was missing in these cases.

Within the lower layer of the atmosphere (up to 3 km) in which dispersion of the most part of radioactive release took place, these data contained from 15 to 25 altitudes at which measurements were performed. Respectively, the spatial resolution of these measurements in vertical direction makes up 20 to 300 m. In meteorological data obtained from Obninsk, the results of radiosounding observation are given only for 5 to 12 points in height, what gives somewhat a lesser resolution in height (the alternative step in height is varying from 200 m to 1500 m).

Under next modeling to determine the values of meteorological parameters in an arbitrary point of the territory for a given value of altitude and moment of time, spatial and temporal interpolation of initial data is being performed.

To describe the input of wet deposition of radioactivity under modeling the complete information of the rainfall gauge network of the Goskomhidromet of Ukraine has been collected, including the data of measurements of 12-hour sums of rainfall at all meteorological stations and posts of Ukraine for the period from April 26th to May 7th, 1986. The rainfall gauge network of the Goskomhidromet of Ukraine in 1986 consisted of 197 meteorological stations and 410 meteorological posts. The characteristic distance between these in flat country is from 20 to 40 km.

Within this period it has been raining almost throughout the Ukraine except the northwest of Volynska region and some places of the flat-country part of western regions of Ukraine (9% of rainfall gauge posts). At more than 50% of stations and posts of the rainfall gauge network, the

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amount of precipitation for this period did not exceed 10 mm. Heavy rains and maximum rainfall rates were noted in the north-east (Kharkiv region) and east (Donets'k region) of Ukraine. It should be stressed that, according to available data, in most cases heavy rain (more 15 mm per 12 hrs) took place in those regions of Ukraine where there was no intensive atmospheric radionuclide contamination caused by the Chernobyl release. However, a final estimate of the contribution of wet deposition into the value of radioactive contamination of the ground surface for different regions of Ukraine, based on quantitative calculations, may be obtained after modeling atmospheric transport and deposition of radioactive release.

2.4. Release specification

2.4.1. *Source term estimations*

The critical point of the Chernobyl release transport modeling is the source term parameterization. It is the largest uncertainty source in the model results due to the lack of direct measurements of radioactive release intensity. A large amount of works is devoted to the estimation of both the total radioactivity amount in the destroyed unit and the amount of radioactivity released into the atmosphere. The first problem may be considered as solved to sufficient degree. Some data of core inventory estimations for ^{137}Cs are shown in Table 2.1 (see UNSCEAR, 2000 also). Results of most these works have been obtained from the analysis of nuclear-physical processes in the reactor core. Despite the difference in initial suppositions and used data the obtained results are quite similar. The same conclusion are made in Pitkevich *et al.* (1993) (the last value in Table 2.1) by averaging of results of 14 original works published. According to that results the standard deviation is about 10 percent for ^{137}Cs while the uncertainties for another radionuclides varied in the range from 5 to 40 percent.

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Table 2.1. Caesium-137 core inventory and total release estimations according different authors

Core inventory (PBq)	Total release estimations (PBq)	Estimated by
280±40	37 (13%) ±50%	Abagyan *et al*. (1986)
300	-	Kirchner and Noack (1988)
310±60	89	Gudiksen *et al*. (1989)
260	-	Begichev *et al.* (1990)
-	74	Izrael *et al*. (1990)
-	70	Ilyin *et al*. (1990)
280	-	Kryshev (1991)
260	-	Borovoi *et al.* (1991)
-	85±26 (33%±10%)	Buzulukov and Dobrynin (1993)
-	89	Borzilov and Klepikova (1993)
280	~ 85	Devell *et al*. (1995)
270±30	85 (30%)	Pitkevich *et al.* (1993)

Values of activity released into the atmosphere may be considered as quite reliable for most radionuclides especially for less volatile ones. The first estimation of ^{137}Cs release value was made in the Soviet report to IAEA (Abagyan *et al*., 1986) as 37 PBq (13% from the cesium inventory) and may be considered as preliminary. The results published lately are quite close and varies in the range 70 - 90 PBq, i.e. about 25-35 percent of total ^{137}Cs amount in the reactor core.

The first scenario of radioactive release dynamics from the 4th unit of Chernobyl power plant during the first days after the accident was suggested in the Soviet report to IAEA (Abagyan *et al*., 1986). According it the released activity was maximal on 26 April (in the first day after the accident) and then decreased next 4 days (Figure 2.1). Later the estimation of total release values was increased in about 2 times (Figure 2.1, 1st version of scenario by Izrael *et al*., 1990) for ^{137}Cs. The next revision of the release dynamics (Figure 2.1, 2nd version of Israel *et al*., 1990) resulted in changes of a qualitative course of release dynamics. Unlike earlier versions the release maximum is

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supposed on third day (28 April). In following days the qualitative trend of the release intensity is the same: decrease of the release intensity up to 30 April – 1 May, and then following increase up to 5 May and sharp reducing of the release intensity after 5 May to values negligibly low comparing with previous ones. The rest of works concerning the estimation of the Chernobyl radioactivity release used more or less close variations of these scenarios (Albergel *et al*., 1988; Haas *et al.*, 1990).

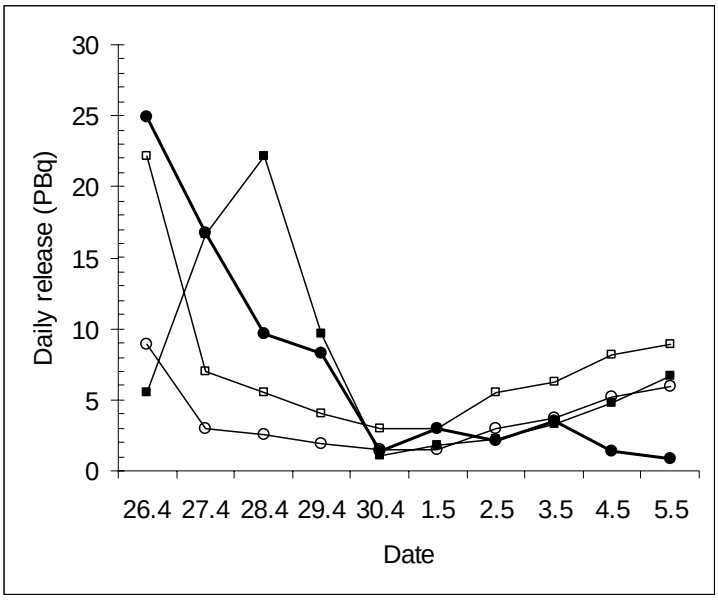

Fig.2.1. Different scenarios of ^{137}Cs daily release from the Chernobyl reactor. Open boxes: Israel *et al*. (1990) – 1st version; solid boxes: Israel *et al*. (1990) – 2nd version; open circles: Abagyan *et al*. (1986); closed circles: present estimations.

2.4.2. *Initial distribution of release over height*

The value of maximal height of radioactive plume initial rise is one of the most crucial parameters under the Chernobyl release transport modeling as follows from preliminary simulations. The time variation of maximal height rise depended on both core temperature variations and local meteorological conditions (wind velocity and temperature stratification in the low part of the atmosphere). According estimations made with using of convective plume model (Talerko, 1990) this value could change considerably in time during the first 10 days of the release. Moreover, a common conception of effective release height for a tall "point" source describes the Chernobyl source inadequately. A convective plume that formed over the accidental unit should be treated as a volume source, which releases a part of radioactive materials through its lateral surface.

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Therefore for this case we should take into account not only value of the maximal height of radioactive plume but the initial distribution of the released radioactivity over height also. Taking into account large amount of calculations the distribution of the released radioactivity intensity was set in 4 grids: at heights 200, 500, 800 and 1200 m. Under next simulation these values were determined with using results obtained from the model Talerko (1990), then used as input data in the LEDI model and afterwards were corrected under the comparison of calculated radioactive contamination fields with measurement results (see next chapter).

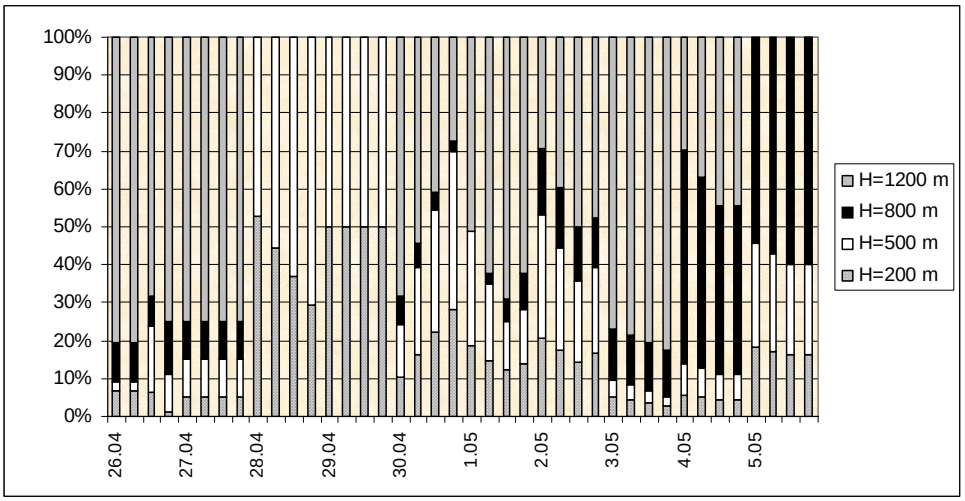

Fig. 2.2. Release relative vertical distribution over heights 200, 500, 800, 1200 m.

2.5. Results and discussion

The scenario of ^{137}Cs daily releases (2nd version by Izrael *et al*., 1990, see Figure 2.1) have been used as an initial approximation for the calculations. The prolonged release in the period from April 26 till May 5 in the calculations have been simulated by a sequence of instant puffs with periodicity 1 hour. The initial amount of activity in each such puff was chosen equals to one 24th of corresponding daily release estimation. Further, the total activity of the release within every hour was arranged between separate puffs with a different initial release height (200, 500, 800 and 1200 m) according chosen time-dependent form of initial release distribution over height. With the help of the LEDI model the dispersion of every puff was simulated and the corresponding field of ^{137}Cs deposition density on the underlying surface was calculated. The total field a radioactive deposition density just as the radionuclide volume concentration were determined as a sum of the contributions

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of all puffs with different release times and different initial heights. Thus, the radioactive release from the destroyed unit within first ten days of the accident was simulated by a set of 960 puffs.

The modeling results have been compared with the measurements data, first of all the results of the soil contamination measurements program carried out in Ukraine during 1986-1991. The comparison between modeling ^{137}Cs cumulative deposition density D_{calc} and the soil contamination measurements data D_{meas} has been made for 12716 settlements in all regions of Ukraine including 4336 settlements in 3 most contaminated Kyiv, Zhytomyr and Chernigiv regions. Using available data the average values of cesium deposition density $\overline{D^i_{calc}}$ and $\overline{D^i_{meas}}$ were estimated for 22 regions of Ukraine (except Kyiv, Zhytomyr and Chernigiv ones). For these most contaminated regions the average values of cesium deposition density were calculated for every district. The function $F = \sum_{i=1}^{N} \left(\overline{D^i_{calc}} - \overline{D^i_{meas}} \right)^2 \Big/ \overline{D^i_{meas}}^2$ was constructed, where N = 22 (number of regions) + 69 (number of districts in 3 regions) = 91. In next modeling steps the activity of each puff is corrected to minimize F and to obtain the best coincidence between the modeling results and ^{137}Cs soil contamination measurements (unlike the initial iteration of calculations, the activity of each puff could vary during a day).

This procedure resulted in detailed ^{137}Cs release scenario (both the activity and the initial height distribution) with the time step 1 hour. The final estimation of daily releases of ^{137}Cs that obtained by summing of puff activities during every day is shown in Figure 2.1. The value of ^{137}Cs total release is estimated to be 72 PBq (26% from core inventory). Final form of release vertical distribution as a function of time obtained under modeling is shown in Figure 2.2 (sums over every 6 h are presented). During periods of 26 - 27 April and 30 April - 4 May the plume height increased up to values of more then 1 km. About 75 - 90 percent of released activity could rise up to these levels. During the period 28 -29 April and to the end of intensive release the maximal plume height decreased to 500 – 800 m.

The general picture of radionuclides transport on regional and global scale are described quite well in literature (Smith and Clark, 1988; Borzilov and Klepikova, 1993; Izrael *et al.*, 1990), so we note only features of radionuclides transport in the territory of Ukraine which resulted in the formation of largest spots of radioactivity contamination field.

During the first hours on 26 April (at night and in the morning) the western and north-western direction of radioactivity transport was predominant within the local zone of the source. Then in the afternoon the main transport direction changed to south-westward (trajectory *a* in

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Figure 2.3 shows the transport of activity released in 12 h 26 April with initial height 1200 m) but further the radioactivity transport turned north-west and activity moved towards Scandinavia. The transport direction variations during this day in time and in height (within the layer 0-1500 m where the main part of released activity moved) caused a wide western trace of the radioactive fallout field in the Ukrainian territory in Zhytomyr, Rivne, Volynska and the part of Kyiv regions (trajectories *a* - *c* in Figure 2.3). By the end of 26 April the main transport direction changed to north-west again in the region of the Chernobyl nuclear power plant and during 27 April the radioactivity moved to the territory of Belorussia. The transport direction during this day and the first half of 28 April was northward and north-eastward toward Chernigiv region of Ukraine and further toward Belorussia and Russia (trajectory *d* in Figure 2.3). We note that these radioactive clouds changed their transport direction over the territory of Russia on 29-30 April and turned to Ukraine again. These radioactive clouds caused the radioactive fallout in the east of Ukraine and then moved to the southern Ukraine and Crimea (trajectories *d – f* in Figure 2.3).

Changing of the atmospheric circulation pattern that began on 28-29 April on the territory of Europe resulted in the essential changing of atmospheric conditions in the local zone of the Chernobyl power plant in the morning 30 April. The large wind velocity shear over height within the low part of the atmosphere resulted in considerable "splitting" of radioactivity plume released this day. So the part of activity which rose to the height 1000 m and more moved south-westward and caused the formation of radioactive fallout spots due to wet deposition of radionuclides in south-western part of Ukraine (Chernivtsy and Khmelnitski regions) (trajectories *a* , *b* in Figure 2.4) whereas the trajectories starting at height 200 m on 30 April (trajectories *e*, *f*) resulted in the beginning of intensive radionuclides fallout to the south from the source (including Kyiv) on 30 April – 1 May that agrees with measurements of ^{137}Cs daily deposition on collectors exposed at network of meteorological stations.

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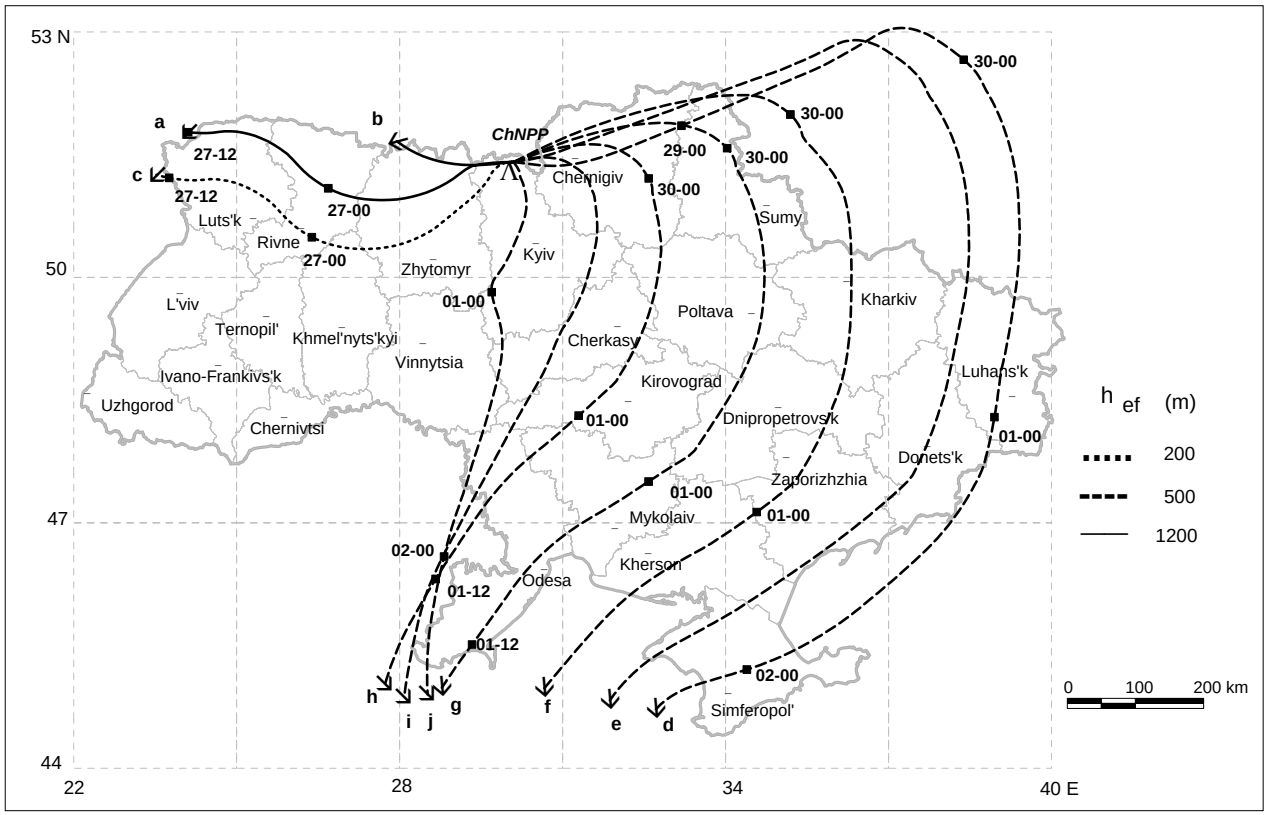

Fig. 2.3. Calculations of radioactivity movement trajectories over the territory of Ukraine for different release moment T_0 (26-29 April 1986) and initial release height h_{ef}:

a) T_0 = 12 hrs 26.04, h_{ef} = 1200 m; b) T_0 = 18 hrs 26.04, h_{ef} = 1200 m;
c) T_0 = 6 hrs 26.04, h_{ef} = 200 m; d) T_0 = 15 hrs 28.04, h_{ef} = 500 m;
e) T_0 = 18 hrs 28.04, h_{ef} = 500 m; f) T_0 = 0 hrs 29.04, h_{ef} = 500 m;
g) T_0 = 3 hrs 29.04, h_{ef} = 500 m; h) T_0 = 6 hrs 29.04, h_{ef} = 500 m;
i) T_0 = 9 hrs 29.04, h_{ef} = 500 m; j) T_0 = 18 hrs 29.04, h_{ef} = 500 m.

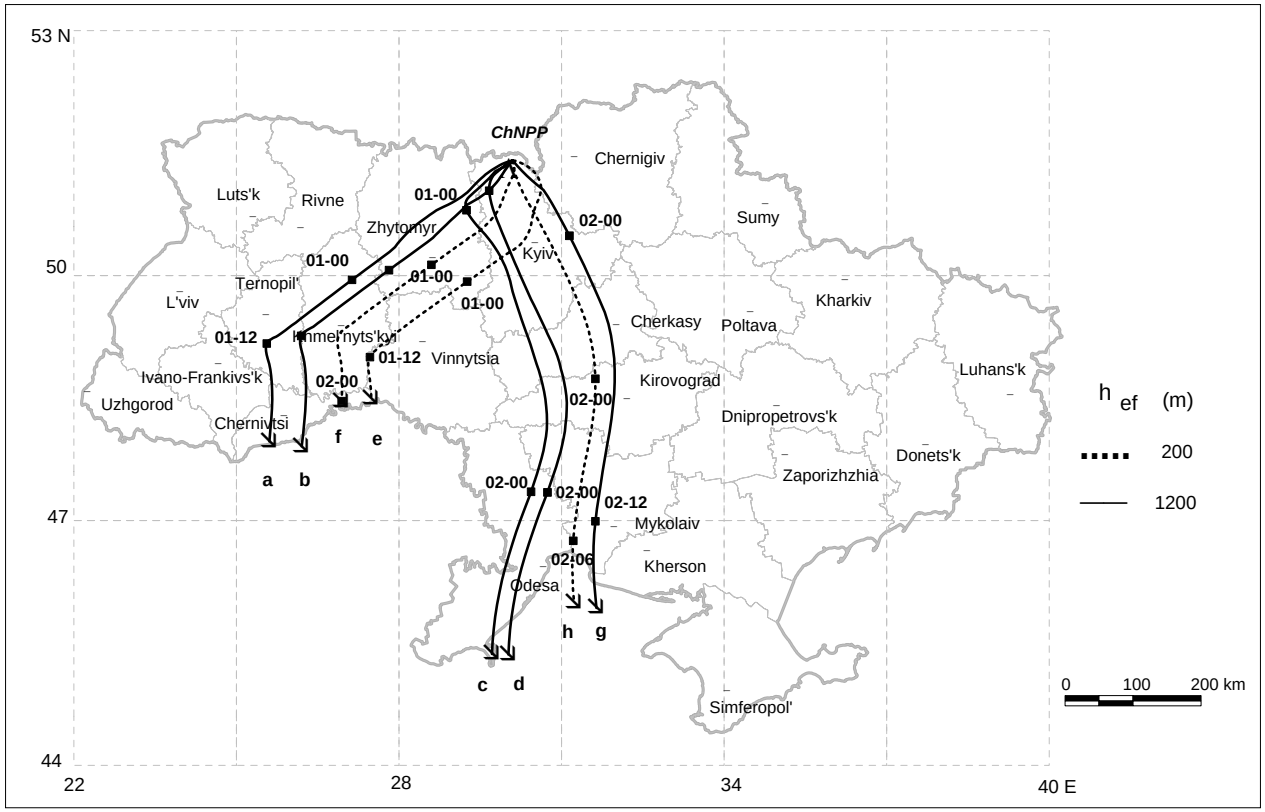

Fig. 2.4. Calculations of radioactivity movement trajectories over the territory of Ukraine for different release moment T_0 (30 April – 1 May 1986) and initial release height h_{ef}:

a) T_0 =	0 hrs 30.04,	h_{ef} =	1200 m;	b) T_0 =	6 hrs 30.04,	h_{ef} =	1200 m;
c) T_0 =	12 hrs 30.04,	h_{ef} =	1200 m;	d) T_0 =	18 hrs 30.04,	h_{ef} =	1200 m;
e) T_0 =	0 hrs 30.04,	h_{ef} =	200 m;	f) T_0 =	6 hrs 30.04,	h_{ef} =	200 m;
g) T_0 =	12 hrs 01.05,	h_{ef} =	1200 m;	h) T_0 =	12 hrs 01.05,	h_{ef} =	200 m.

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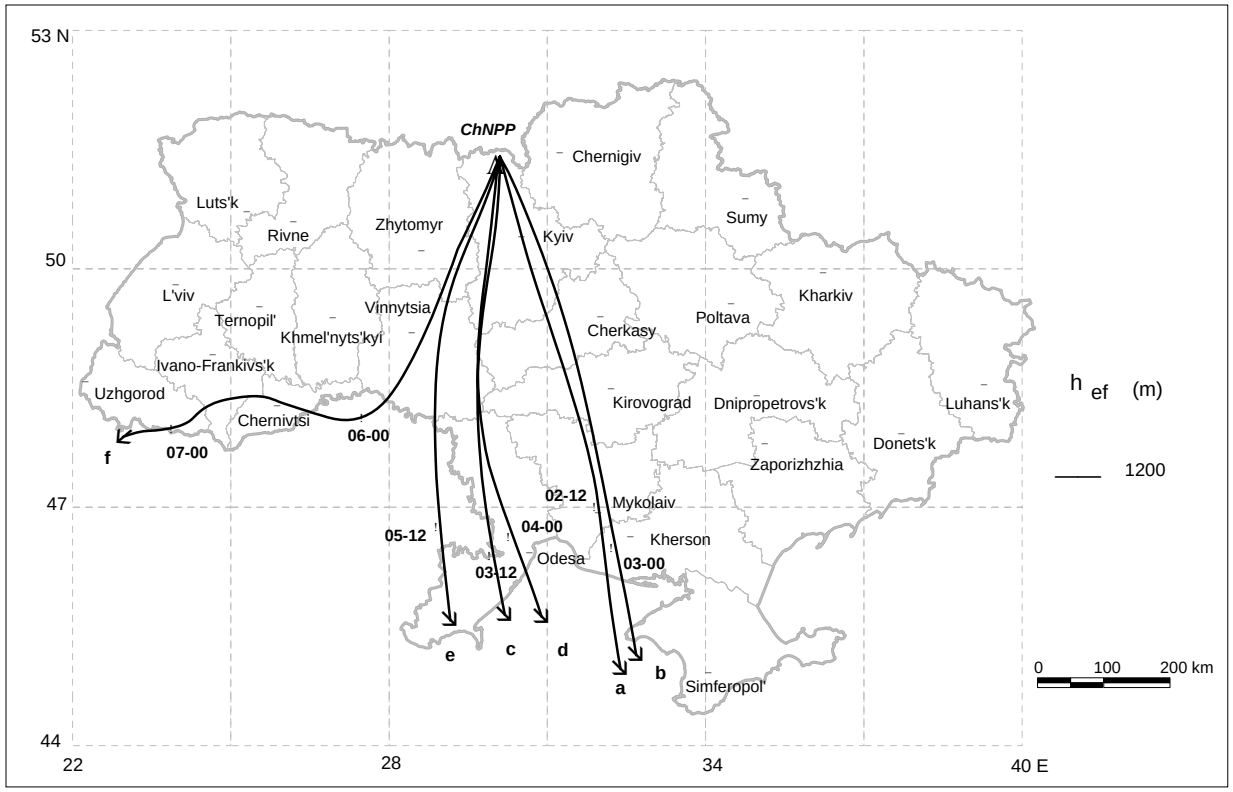

Fig. 2.5. Calculations of radioactivity movement trajectories over the territory of Ukraine for different release moment T_0 (2 – 4 May 1986) and initial release height h_{ef}:

a) T_0 = 0 hrs 02.05, h_{ef} = 1200 m; b) T_0 = 12 hrs 02.05, h_{ef} = 1200 m;
c) T_0 = 0 hrs 03.05, h_{ef} = 1200 m; d) T_0 = 12 hrs 03.05, h_{ef} = 1200 m;
e) T_0 = 0 hrs 04.05, h_{ef} = 1200 m; f) T_0 = 12 hrs 04.05, h_{ef} = 1200 m.

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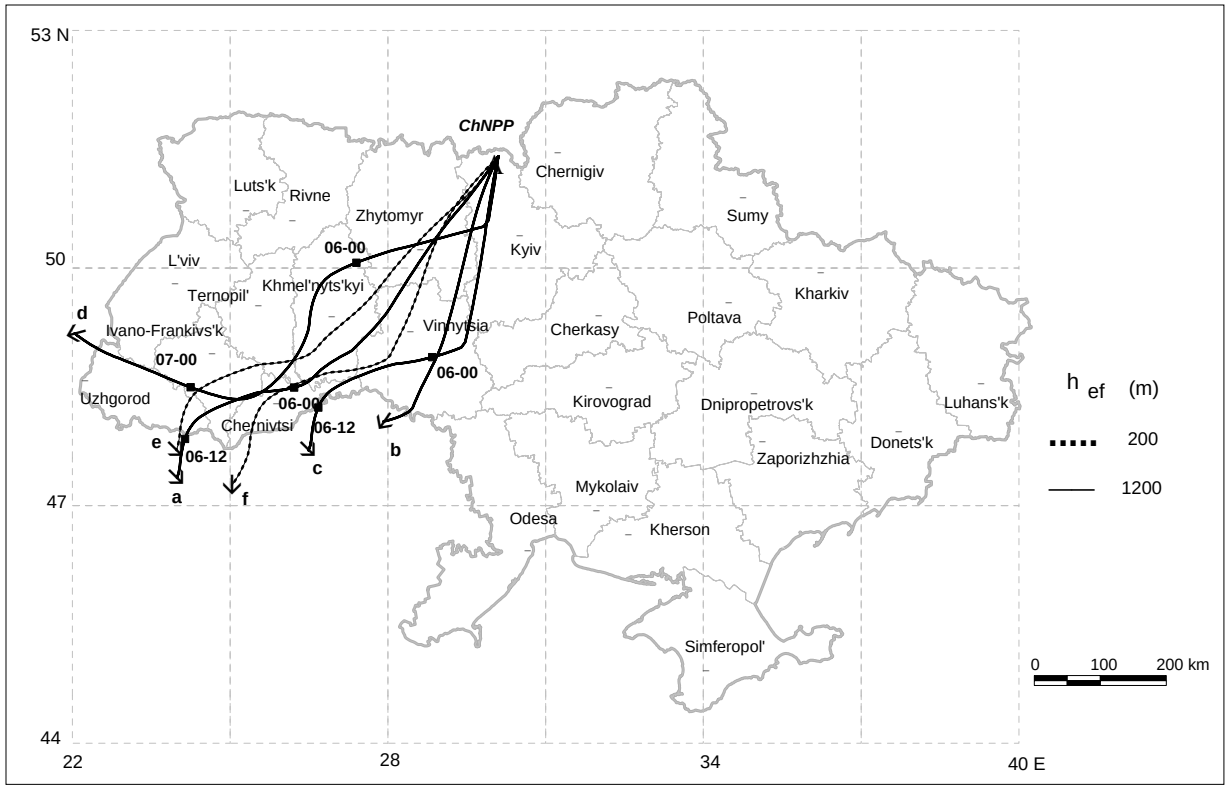

Fig. 2.6. Calculations of radioactivity movement trajectories over the territory of Ukraine for different release moment T_0 (5 May 1986) and initial release height h_{ef}:

a) T_0 = 0 hrs 05.05, h_{ef} = 1200 m; b) T_0 = 6 hrs 05.05, h_{ef} = 1200 m;

c) T_0 = 12 hrs 05.05, h_{ef} = 1200 m; d) T_0 = 18 hrs 05.05, h_{ef} = 1200 m;

e) T_0 = 0 hrs 05.05, h_{ef} = 200 m; f) T_0 = 6 hrs 05.05, h_{ef} = 200 m.

The analysis of rain measurement data and the calculation results shows that the radioactive deposition field in the central part of Ukraine formed mainly due to dry deposition on the underlying surface. The contribution of wet deposition in this region was negligibly low. Nevertheless, according to soil activity measurement data there are several spots of relatively large (over 37 kBq m^{-2} of ^{137}Cs) deposition in Cherkasy region and the south part of Kyiv region. According to the simulation results the main reason of formation of such large-scale radioactive deposition spot at the distances 200-400 km from the source was an influence of changed meteorological conditions of release transport (due to daily variations of parameters of the planetary boundary layer where the main part of release moved). At night in the conditions of shallow stable boundary layer the turbulent exchange is low, so deposition of pollutant released from an elevated source was small in the near zone of the source. Therefore for the release from elevated source with the maximal height which exceeds the nocturnal boundary layer height (with typical values about 100 - 300 m) the main part of released material rose over the layer of turbulent mixing. In this situation the released material was transported by horizontal wind without considerable vertical mixing. So the high-level releases during several hours in night could result in the pollutant transport to the distances up to several dozens or even hundreds kilometers (depending on the wind speed at the levels of transport). As a consequence of daily variations of turbulent parameters of the boundary layer its height and the turbulence intensity increase in the morning and in the daytime due to a warming up of the surface. As a result of increasing of the planetary boundary layer height (it could reach the value 1000 – 2000 m) in some moment the boundary layer “captures” the elevated release and therefore the pollutant are involved in the intensive vertical turbulent exchange. Some time later the pollutant reaches the ground resulting in the formation of deposition spot far away from the source.

We note that the mechanism of the deposition spots formation at the mesoscale distances due to the combination of diurnal variations of the transport conditions and the high-elevated prolonged release source is quite obvious in principle. But just Chernobyl release with a large maximal rise height apparently became the first and for the present the only case in a reality. On the other hand, any possible heavy radiation-thermal accident at night could result in analogous consequences. In this situation the radiological measurements in the near zone of the source couldn’t ensure the control in far away regions.

During next days of the accident (from 1 to 4 May) the predominant transport direction continued to change clockwise, and the vertical shear of wind direction was quite small. It caused a relatively narrow trace of radioactive deposition (Figure 2.5) in the south. The activity released

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during 5 May caused the subsequent formation of large-scale deposition spot in the western and south-western part of Ukraine (Vinnytsia, Khmel'nyts'kyi and Chernivtsi regions) (Figure 2.6). By to 7 May the formation of south trace of radioactive deposition finished in the territory of Ukraine.

Table 2.2 presents the distribution of calculated and measured ^{137}Cs deposition density ratios averaged over settlements for every region. This ratio *R* doesn't differs more than in 1.5 times for 61.5 percent of total number of settlements that could be considered as rather good agreement. For 180 settlements (1.5%) this ratio is greater 5. Estimations of total modeled and measured deposition density ratio for each region are presented in last column of Table 2.2. It varies from 0.75 to 1.15 with mean value 0.91 for all Ukraine and 0.92 for 3 most contaminated regions.

Otherwise, the radioactive contamination in every settlement is described not only mean value but real distribution of measurement results. The standard deviation (STD) of ^{137}Cs soil contamination has been calculated for every settlement using available individual measurement data. The value of STD averaged over all settlements is 14.0 Bq m^{-2} (19.1 Bq m^{-2} for settlements of 3 most contaminated Kyiv, Chernigiv and Zhytomyr regions; 7.7 Bq m^{-2} for the rest of Ukraine). The ratio of STD to soil contamination was calculated for every settlement. This value averaged over all settlements is 0.44 (0.45 for 3 most contaminated regions and 0.43 for the rest of Ukraine). For 106 settlements of Ukraine (1.4%) the value of STD is larger than mean soil contamination value. So the another criteria of $| D_{calc} - D_{measur} | <$ STD have been chosen to estimate the agreement between modeling and measurements for every inhabitant place. According to Table 2.3 this criteria yields roughly the same result – the model results are quite good for about two thirds of total settlement number. The results of comparison for all settlements over Ukraine are similar.

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Table 2.2. Comparison between model results and measurements of ^{137}Cs deposition density in the settlements in Ukraine.

Region	Number of settlements (where measurement data are available)	Percentage of settlements with given deposition ratio $R=D_{calc} / D_{measur}$							Ratio of modeling and measured total deposit
		$R<0.2$	$0.2<R<0.33$	$0.33<R<0.67$	$0.67<R<1.5$	$1.5<R<3$	$3<R<5$	$R>5$	
Vinnytsia	616	2.8	3.6	14.3	51.8	24.2	2.9	0.5	0.72
Volynska (Rivne)	346	0	0.9	12.4	63.3	22.0	1.4	0	1.05
Lugans'k	278	0.7	1.1	19.8	74.8	3.6	0	0	0.93
Dnipropetrovs'k	280	0.4	4.3	23.2	63.6	7.1	1.1	0.4	0.75
Donets'k	315	0.3	2.2	23.5	69.8	3.2	0.6	0.3	0.83
Zhytomyr	1666	0.3	1.0	9.7	63.6	24.4	0.9	0.2	0.93
Zakarpatska (Uzhgorod)	232	0.4	3.0	21.1	69.0	6.5	0	0	1.00
Zaporizhzhia	240	0	0.8	14.2	78.8	6.3	0	0	0.94
Ivano-Frankivs'k	346	0.3	3.2	22.0	62.4	10.4	1.2	0.6	0.85
Kyiv	1217	2.5	2.8	17.7	57.1	18.2	1.6	0.2	0.91
Kirovograd	328	0.3	0.6	13.7	68.0	17.4	0	0	1.01
Crimea	296	0.3	0.7	11.1	81.1	6.8	0	0	0.95
L'viv	548	2.4	1.6	25.2	58.9	11.5	0.4	0	0.87
Mykolaiv	249	0.4	0	6.0	72.3	20.5	0.4	0.4	1.06
Odesa	391	0.3	1.0	6.9	62.7	27.1	1.8	0.3	1.15
Poltava	384	0	0	9.1	87.8	3.1	0	0	1.04
Rivne	316	0	0.9	13.3	59.5	25.6	0.6	0	1.02
Sumy	353	0.6	3.4	21.5	55.8	12.7	4.5	1.4	1.11
Ternopil'	390	0.5	3.8	13.8	50.5	29.2	2.1	0	0.89
Kharkiv	370	0	0	14.1	80.8	4.9	0.3	0	0.97
Kherson	266	0	0	8.6	85.3	4.9	1.1	0	1.02
Khmel'nyts'kyi	473	2.3	2.7	10.6	54.1	27.3	2.5	0.4	0.85
Cherkasy	459	1.7	4.6	17.0	52.5	21.1	2.8	0.2	0.73
Chernivtsi	220	1.8	4.5	15.0	61.8	16.4	0.5	0	0.72
Chernigiv	1525	3.2	3.2	15.7	45.4	26.7	4.8	1.0	0.92
3 most contaminated (Kyiv, Chernigiv, Zhytomyr) regions	4408	1.9	2.2	14.0	55.5	23.5	2.4	0.5	0.92
Ukraine (total)	12104	1.2	2.1	14.9	61.5	18.2	1.7	0.3	0.91

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Table 2.3. Comparison between model results and measurements of ^{137}Cs deposition density in the settlements in Ukraine according the verification criteria $| D_{calc} - D_{measur} | < STD$.

Region	Total number of settlements	Percentage of settlements		
		Low	Within interval	High
Vinnytsia	413	16	56	28
Volynska (Lutsk)	336	5	68	27
Lugans'k	83	11	86	4
Dnipropetrovs'k	64	22	66	13
Donets'k	45	16	80	4
Zhytomyr	1633	6	65	29
Zakarpatska (Uzhgorod)	25	12	84	4
Zaporizhzhia	4	0	100	0
Ivano-Frankivs'k	159	22	58	19
Kyiv	1135	9	73	19
Kirovograd	87	17	67	16
Crimea	45	7	80	13
L'viv	528	23	62	15
Mykolaiv	55	4	85	11
Odesa	93	5	70	25
Poltava	4	0	100	0
Rivne	310	8	63	29
Sumy	76	22	49	29
Ternopil'	209	22	47	31
Kharkiv	51	0	90	10
Kherson	1	0	100	0
Khmel'nyts'kyi	216	20	44	36
Cherkasy	375	14	59	27
Chernivtsi	217	17	59	24
Chernigiv	1513	16	49	35
3 most contaminated regions	4281	10	61	28
Ukraine (total)	7677	13	61	26

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The map of calculated ^{137}Cs deposition density field in Ukraine is presented in Figure 2.7. The agreement of computed deposition density field with soil contamination measurement data is rather good. The formation of main deposition spots in the territory of Ukraine have been reconstructed under modeling including western and north-eastern arms of deposition field just as the ^{137}Cs spots in central and southern part (Kyiv, Cherkasy, Vinnytsia, Khmel'nyts'kyi regions).

Another set of measurement data used for the model results verification is measurements of ^{137}Cs daily deposition on collectors exposed at network of meteorological stations in April-May 1986. Unfortunately, there were only two measurement points in the regions of intensive radioactive contamination - Kyiv and Baryshevka (Kyiv region). Table 2.4 presents the dynamics of daily deposition values according modeling and measurements. The modeling results agree rather well for both measurement points for both total deposition values and for every day of intensive fallout. For Kyiv the cumulative ^{137}Cs deposition density is 26.5 kBq m^{-2} according modeling, 26.0 kBq m^{-2} from soil measurement and 19.7 kBq m^{-2} obtained by summing of results daily measurements of deposition at meteorological station. For Baryshevka these results are 8.7 kBq m^{-2} (model), 8.7 kBq m^{-2} (soil measurement) and 10.7 kBq m^{-2} (deposition at station).

Table 2.4. Modeled and measured ^{137}Cs daily deposition density in Kyiv and Baryshevka (kBq m^{-2})

		26-27.04	27-28.04	28-29.04	29-30.04	30.04-01.05	01-02.05	02-03.05	03-04.05	04-05.05	Total
Kyiv	Calc.	0	0	0	0	4.3	13.5	7.1	1.1	0	26.0
	Measur.	0.025	0.037	0.04	0.13	3.6	11.9	3.4	0.42	0.16	19.7
Baryshevka	Calc.	0	0	0	0	3.9	3.6	1.1	0.12	0	8.7
	Measur.	0.022	0.009	0.014	0.074	7.5	2.6	0.23	0.13	0.10	10.7

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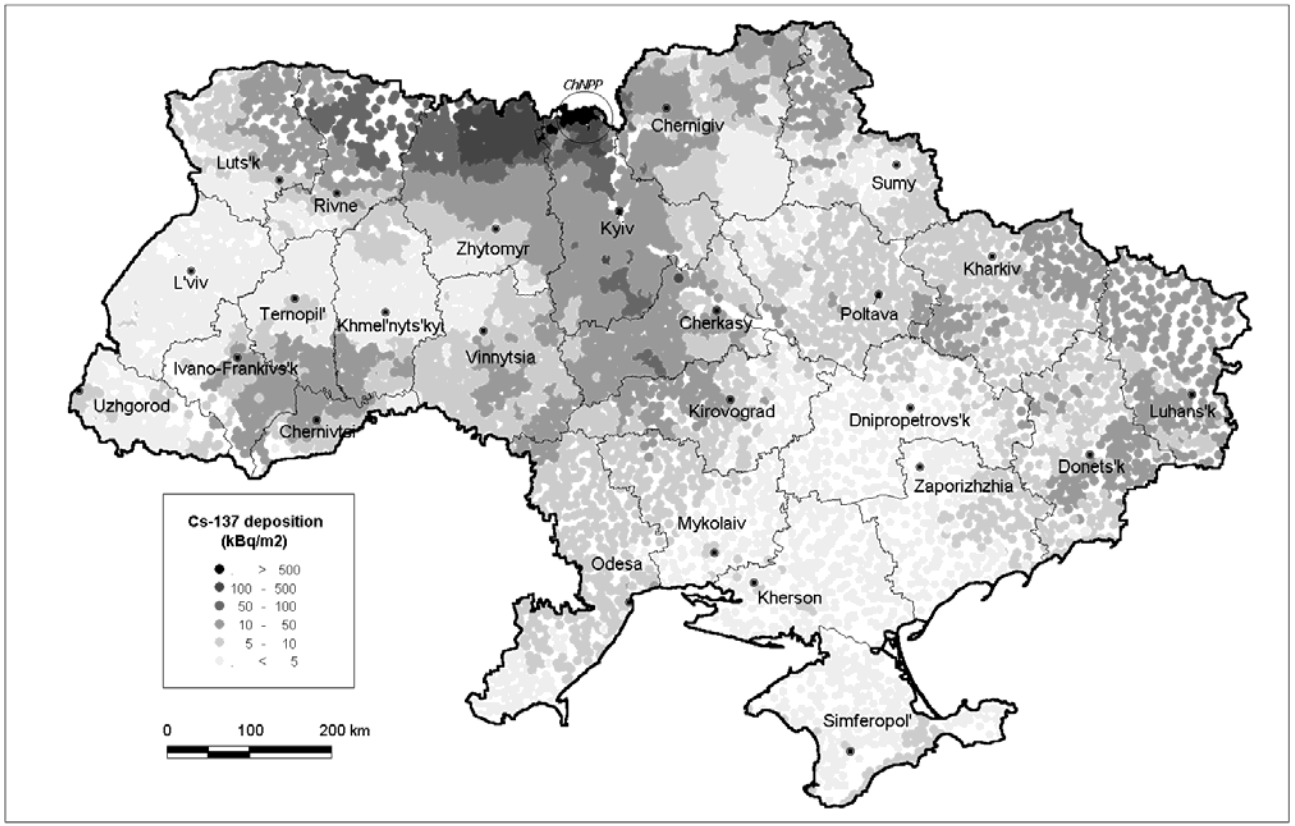

Fig. 2.7. Calculated field of ^{137}Cs surface ground deposition in Ukraine.

2.6. Conclusions

It is worth noticing that despite a great number of fulfilled investigations on the Chernobyl release transport the perfect and non-contradictory picture of the radioactivity contamination formation for different radionuclides on different space scales haven't constructed yet. It can be explained by the extreme complexity of this problem including a great number of uncertain parameters, especially of the source term. Despite it the simulations using atmospheric transport models for different space scales could be useful (and only possible in some cases) way for the reconstruction of environmental radioactive contamination and the impact on peoples' health caused by the Chernobyl accident.

The comparison of modeling results and measurements data for ^{137}Cs showed the validity of used approach for the reconstruction of the Chernobyl release atmospheric transport over Ukraine. The simulation with the aid of LEDI model and available meteorological data just as unavoidable additional hypothesis about the source term parameters and the following comparison with measurement data enables to obtain the realistic picture of radioactivity transport and deposition in the territory of Ukraine on the regional scale. The initial scenario of the activity releases from the accidental unit of the Chernobyl NPP have been corrected and worked out in detail. So it gave the ground to apply this approach for the reconstruction of the ^{131}I contamination dynamics in Ukraine. The calculated airborne ^{131}I concentration and ground deposition density fields could be used as the database for thyroid dose reconstruction for inhabitants of radioactive contaminated regions in Ukraine.

3. MODELING OF ^{131}I RADIOACTIVE ATMOSHERIC TRANSPORT IN UKRAINE CAUSED BY THE CHERNOBYL ACCIDENT

The task of thyroid dose reconstruction for inhabitants of radioactive contaminated regions after the Chernobyl accident is still of special importance (Likhtarev *et al*., 1999) because of the observed increase in the number of thyroid cancer in these regions of Ukraine, Belorussia and Russia. Unfortunately, the lack of ^{131}I soil contamination data (especially for the territory of Ukraine) does not enable to carry out such reconstruction using direct measurements. Besides it, the results of reconstruction the thyroid dose are considerably dependent on time-variable air-borne concentration values and especially on times of the activity arrival and the finishing of fallout which

are specific for every settlement.

3.1. Source term estimation

The first assessment of ^{131}I inventory in the accidental reactor core was 1300 PBq (decay-corrected to 6 May 1986) (IAEA, 1986) and then the more reliable estimation 3200 PBq was obtained (Begichev *et al*., 1990). According to it the estimation of total ^{131}I release was changed from 260 PBq (IAEA, 1986) to 1760 PBq (estimation of total release during the coarse of the accident) (UNSCARE, 2000). According to Pitkevich *et al.* (1993) the standard deviation of ^{131}I release estimation is about 10 percent.

Table 3.1. Different scenarios of ^{131}I daily release (in PBq) from the Chernobyl nuclear power plant.

Date	Izrael *et al*. (1990) – the older version	Izrael *et al*. (1990) – the newer version	Haas	Borzilov, Klepikova (1993)	UNSCEAR (2000)	Our estimations
26 April	192	33	149	241	704	339
27 April	56	111	46	70	204	227
28 April	41	152	42	32	150	130
29 April	28	54	33	23	102	113
30 April	19	11	20	26	69	23
1 May	17	11	20		62	45
2 May	28	15	42		102	29
3 May	30	21	136		107	45
4 May	35	31	64		130	16
5 May	36	44	120		130	9
Total release	480	482	670	392 (during first 5 days)	1760	975

The several scenarios of radioactive release dynamics from the 4th unit of Chernobyl power plant during the first days after the accident is shown in Table 3.1 including two versions of the release estimations made by Izrael (1990), two scenarios used in atmospheric transport modeling

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works (Haas, 1990) and Borzilov, Klepikova (1996) and the ^{131}I daily releases scenario from UNSCEAR (2000). The last column in Table 3.1 is our estimation of ^{131}I release. To construct it the next assumption have been used: 1) the $^{131}I/^{137}Cs$ deposition density ratio in the nearest zone of the source was taken according to Muck (2002) equal to 19.08 for western and north-eastern trace of radioactive fallout field (formed in the main due to releases during first 4 days – from 26 April to 29 April); 2) the correction factor 1.7 is applied for $^{131}I/^{137}Cs$ deposition density ratio in the nearest zone for southern trace (formed in the main due to releases during the period from 30 April to 5 May); 3) the ratio of $^{131}I/^{137}Cs$ deposition velocity is taken to be 1.4 according the assessments of radionuclides deposition velocities based on measurements in Kyiv (Izrael *et al*., 1990). To meet these assumptions the ratio $^{131}I/^{137}Cs$ was taken equals to 13 for first 6 days after the accident (up to 1 May), then these ratio decreased to 12.7 on 2 May, 11.7 on 3 May, 10.7 on 4 May and 9.8 on 5 May (taking into account the radioactive decay).

The total ^{131}I release is estimated to be 975 PBq or 30.5% of the core inventory. The corresponding value decay corrected to 26 April is 1170 PBq.

The time dependence of iodine release is similar for the most of scenarios except version 2 of Izrael (1990) suggested the release maximum on 28 April. For the rest the largest release was on 26 April, then decreased and the second maximum was assessed on 3-5 May. It should be noted that all scenarios based on the comparison of activity atmospheric transport and deposition modeling suggest much less values of iodine release in the comparison with the UNSCEAR scenario which could be considered as the most reliable one. This difference is the largest for assessments of the release during the first day. The possible reason is that the greater part of modeling works use the available data of activity measurements in soil and air for the reconstruction of source term parameters including daily releases estimation. But the radioactive contamination on local or regional scale has been formed due to not total radioactive release but only it's part which dispersed in lower part of the atmosphere and could deposit on the underlying surface during the first days of the accident. On the other hand, another part of activity from the accidental unit could raise to much higher levels, resulting in radioactive contamination of heights about several kilometers (Kownacka, Jaworowski, 1987) and didn't affect to radioactive contamination of ground level air near the source. So this difference between total release estimation in UNSCEAR (2000) and another estimations (including ours) may be interpreted as the part of activity injected in troposphere and lower stratosphere and dispersed at large distances from the Chernobyl nuclear power plant.

As for the cesium modeling the Chernobyl source is treated as a volume one due to the

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formation a convective plume over the destroyed unit and the release of a part of radioactive materials through its lateral surface. The parameters of this volume source (maximal height and initial vertical distribution of release intensity) were considered under the modeling of radionuclides atmospheric transport as functions of time during the period of intensive releases (Talerko, 2004). The radioactivity release intensity was set in 4 grids: at heights 200, 500, 800 and 1200 m. Daily averaged relative ^{131}I vertical distribution release intensity used in calculation are shown in Table 3.2.

Table 3.2. Daily averaged relative ^{131}I vertical distribution release intensity used in calculation.

Date	200 m	500 m	800 m	1200 m
26.04	0.05	0.10	0.10	0.75
27.04	0.05	0.10	0.10	0.75
28.04	0.30	0.20	0.10	0.40
29.04	0.32	0.47	0.02	0.19
30.04	0.22	0.37	0.16	0.26
1.05	0.37	0.23	0.07	0.32
2.05	0.40	0.35	0.09	0.15
3.05	0.04	0.07	0.12	0.76
4.05	0.08	0.12	0.55	0.24
5.05	0.27	0.46	0.27	0.00

3.2. Iodine-131 dry deposition velocity

Radioiodine in the Chernobyl release occurred in three different physical-chemical forms: aerosol, molecular and organic ones. It creates additional problems for measurements of iodine volume concentration because the great part of radioactivity sampling in the USSR during the initial period of the accident were made with using of gauze collectors and measured the aerosol part of

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the release only. The rare results of different iodine forms measurements on the territory of the former USSR are known. Styro *et al*. (1992) reported that the percentage of the aerosol, molecular and organic forms was 24:22:54 on 30 April 1986 (the day of maximal concentration of ^{131}I in the air) and 35:14:51 on the average from 30 April to 10 May using the data of measurements carried out in Vilnius (Lithuania). Unfortunately, transformation of these physical-chemical forms is still poorly investigated. During the atmospheric transport the transformation between these physical-chemical forms occurs. Elementary iodine released from the reactor interacts with condensation nucleus of the atmospheric air and adsorbed on them. On the other hand sublimation of deposited iodine occurred that complicated the interpretation of measurement results and decreased the reliability of obtained results. Since the constants of these transformations in the natural conditions are unknown there are no data for the reconstruction of the fractions of each iodine forms in the initial release. Because of these problems it seems unfounded in the modeling task to divide total ^{131}I release between these three forms and to examine separately the transport of each form and the transformation between different forms. So, to our mind, the only way under iodine atmospheric transport modeling is to consider the iodine isotopes of all forms as a uniform substance. From the point of modeling of atmospheric transport this assumption is quite acceptable because the typical size of transported aerosol particles doesn't exceed several micrometers, and the gravitational sedimentation of aerosol particles may be neglected. But another great problem is the great difference between the values of dry deposition of the iodine forms. The results of laboratory and natural measurements of radioiodine aerosol deposition velocity gives the values varying in the range from 0.1 cm s^{-1} to 1 cm s^{-1} (Ilyin *et al*., 1972; Nickolson, 1988) depending on the particle size, the underlying surface characteristics and meteorological conditions. The dry deposition of ^{131}I on the underlying surface in the organic form is estimated to be 0.01 cm s^{-1}, whereas the dry deposition velocity of elementary iodine is estimated to be 2 cm s^{-1} on dry grass and 3 cm s^{-1} on wet grass (Makhon'ko *et al*., 1996).

Comparison of works on atmospheric transport modeling of the Chernobyl release reveals the tendency to decreasing of the dry deposition velocity used in models when increasing space scale of modeling region. Israel *et al*. (1987) obtained parameters of size distribution of the Chernobyl release aerosol particles from comparison of the results of atmospheric transport modeling with the data of gamma-dose measurements. Assumed log-normal distribution they calculated the values of particle median diameter ξ and the logarithm of the geometric standard deviation σ. It was obtained that ξ=50 μm and of σ=0.25 for mesoscale distances (up to 100 km), ξ=20 μm, σ=0.4 for regional scale (up to 400 km) and ξ=5 μm, σ=0.25 for the problem of long-

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distance transport modeling over Europe. For these estimations the dry deposition velocities for particles median diameters equal 19, 3 and 0.2 cm s^{-1} accordingly. The modeling works on the reconstruction of ^{131}I and ^{137}Cs deposition on the territory of the former USSR (Pitkevich *et al*., 1994; Borzilov *et al.*, 1988) used the value of V_g = 1 cm s^{-1}. From another side, the typical values of ^{131}I dry deposition V_g=0.1 - 0.3 cm s^{-1} has been used in most modeling works concerning the long-range transport of the Chernobyl release in the Western Europe (Albergel *et al*., 1988; Izrael *et al*., 1990). The last values are seems well grounded for the problem of the Chernobyl release deposition on the distances over 1000 km from the source. However it's necessary to note that presumably only thin-dispersed part of the initial release was transported as far as such large distances. Furthermore, the part of iodine release in the elementary form was small at large distances as deposited earlier. So the situation may be quite different in the close region near the Chernobyl power plant the same way as on meso- and regional scales (up to 200-500 km from the source). Both the elementary form contribution and the relatively large part of aerosol size distribution can result in considerable larger deposition values.

The results obtained for the Chernobyl release suggest the same tendency of decreasing of dry deposition velocity with increasing of the distance from the source. A lot of measurements of the air activity and deposition during the first days after the accident were made in the territory of Western Europe. It yield a deposition velocity of about 0.04 - 0.2 cm s^{-1} for ^{137}Cs deposition to grass (Nickolson, 1988). The radionuclides dry deposition velocity in the closest region of the Chernobyl power plant was calculated by Israel *et al*. (1990) using data of daily measurements of volume activity and deposition values obtained in the initial period of radioactive releases at the meteorological stations in Baryshevka (settlement in Kyiv region at 140 km to the east-south from the source) and in Minsk (Belarus, 330 km from the source). These dry deposition values for main radionuclides (including aerosol part of ^{131}I) are in the range from 0.4 cm s^{-1} to 1.8 cm s^{-1} that are essentially large than ones obtained from the measurements in the Western Europe.

Makhon'ko *et al.* (1996) suggested the value of the ^{131}I deposition velocity V_g=0.8 cm s^{-1} based on the data of the measurements of radioactive iodine made in Obninsk (Russia) on 30.04 – 01.05.1986. As it was outlined, this value is very close to one obtained for the global radioactive deposition V_g=0.7 cm s^{-1}. Besides it was estimated the dry deposition velocity using the above-mentioned ratio between different physical-chemical forms measured in Vilnius and dry deposition values for each forms. The value V_g=0.6 cm s^{-1} was obtained for deposition on dry grass and V_g=0.9 cm s^{-1} for deposition on wet grass. Taking these results in consideration we consider the transport of ^{131}I of the Chernobyl release over the territory of Ukraine as uniform substance without division

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between different forms and use the averaged ^{131}I dry deposition velocity V_g=0.7 cm s^{-1}.

3.3. Numerical simulation

The model of atmospheric pollutant transport LEDI was used for the reconstruction of dynamics of ^{131}I air and underlying surface contamination of the territory of Ukraine during an initial phase of the Chernobyl accident. The procedure of ^{131}I transport and deposition was similar to one used for ^{137}Cs. As the input meteorological data for modeling the data of radiosounding measurements of the atmosphere performed by a network of aerological stations in the former USSR were used. The data of measurements performed 4 times a day during 25 April to 7 May 1986 at 9 stations in Ukraine and 2 nearest stations in Belarus and Russia were used. Under modeling of radioactive transport to determine the values of meteorological parameters of atmosphere in an arbitrary point of the territory for a given value of altitude and moment of time, spatial and temporal interpolation of initial data was performed. To describe the input of wet deposition of ^{131}I under modeling the data of measurements of 12-hour sums of rainfall at 197 meteorological stations and 410 meteorological posts in Ukraine for the period from April 26th to May 7th, 1986 were used.

The prolonged release in the period from April 26 till May 5 in the calculations has been simulated by a sequence of instant puffs with periodicity 1 hour. The amount of activity in each such puff was selected so that the summarized ^{131}I activity in all puffs for every day corresponded to the chosen release scenario (Table 3.1). The summarized activity of the release within every hour was arranged between separate puffs with a different initial release height (200, 500, 800 and 1200 m) according chosen form of initial distribution of release over height (Table 3.2). So the total fields of ^{131}I deposition and volume concentration were calculated as a sum of the contributions of all 960 puffs with different release times and different initial heights.

One of the main task of this work was to create the database of calculated ^{131}I activity in air and ground during period from 26 April to 7 May 1986 in every inhabitant place in Ukraine to provide with necessary information the retrospective dosimetry studies of the reconstruction of thyroid doses of the Ukrainian population caused by the Chernobyl accident. So values of daily deposition and time-averaged air volume concentration were calculated in 12715 inhabitant places of Ukraine.

The atmospheric transport model LEDI and used meteorological data enable to obtain the relatively large-scale (with scale of about several dozens of kilometers) features of fields of

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radioactive contamination of Ukraine. Though, as the results of radioactive contamination measurements show, there is a considerable heterogeneity of deposition field with scale about 1 km. This variability may be explained different reasons: impact of local features of underlying surface which don't take into account under simulation (horizontal resolution of used set of topography and landuse data was 10 km), different characteristics of deposited material (e.g., variability of relative input of different physical and chemical forms of radioiodine), a possible input of wet deposition caused by local rains which haven't been detected with the rainfall gauge network etc. So to describe the local variability of radioactive deposition field we used the approach similar to one used by Pitkevich *et al*. (1994) and named by them as "the method of local effective depositions". According it, the correction factor is introduced for every inhabitant place to come into agreement the modeling results with measured values. This factor may be calculated as the ratio between calculated D_{calc} and measured D_{meas} integral ^{137}Cs deposition densities. As stated in Chapter 2 this values doesn't differs more than in 1.5 times for 61.5 percent of total number of settlements, whereas for 1.5% settlements this ratio is greater 5. This value may treated as a measure of difference between a local deposition and smoothed calculated deposition field. We supposed that these local differences are the same as for ^{137}Cs as for ^{131}I deposition fields. So the correction factor of "local effective depositions" is equal to $R = D_{meas}^{Cs-137} / D_{calc}^{Cs-137}$. It was calculated for every inhabitant place and applying to results of ^{131}I deposition modeling results. The final value of iodine deposition density is equal to

$$D^{I-131} = R \cdot D_{calc}^{I-131}.$$

This correction is applied to both daily deposition and cumulative deposition density values.

3.4. Results and discussion

The general pattern of ^{131}I air and ground contamination fields formation in the territory of Ukraine is similar to ^{137}Cs one and includes next stages:

1) Formation of the western trace of contamination field (including Zhytomyr, Rivne, Volyn regions) during 26 and 27 April (Figure 3.1-3.2);
2) Changing the main transport direction to the north and north-east to Belarus and Russia (Figure 3.3);
3) Clockwise transport direction rotation resulted in the beginning of radioactive contamination of central (including Kyiv), south-western (including Chernivtsi and

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Hmel'nitskiy regions) and eastern parts of Ukraine by 30 April and 1 May (Figure 3.4-3.5);

4) Next formation of southern part of radioactive contamination field during 2-6 May 1986 (Figure 3.6-3.10);

5) Finishing of radioactive deposition formation in the territory of Ukraine by to 7 May (Figure 3.11-3.12).

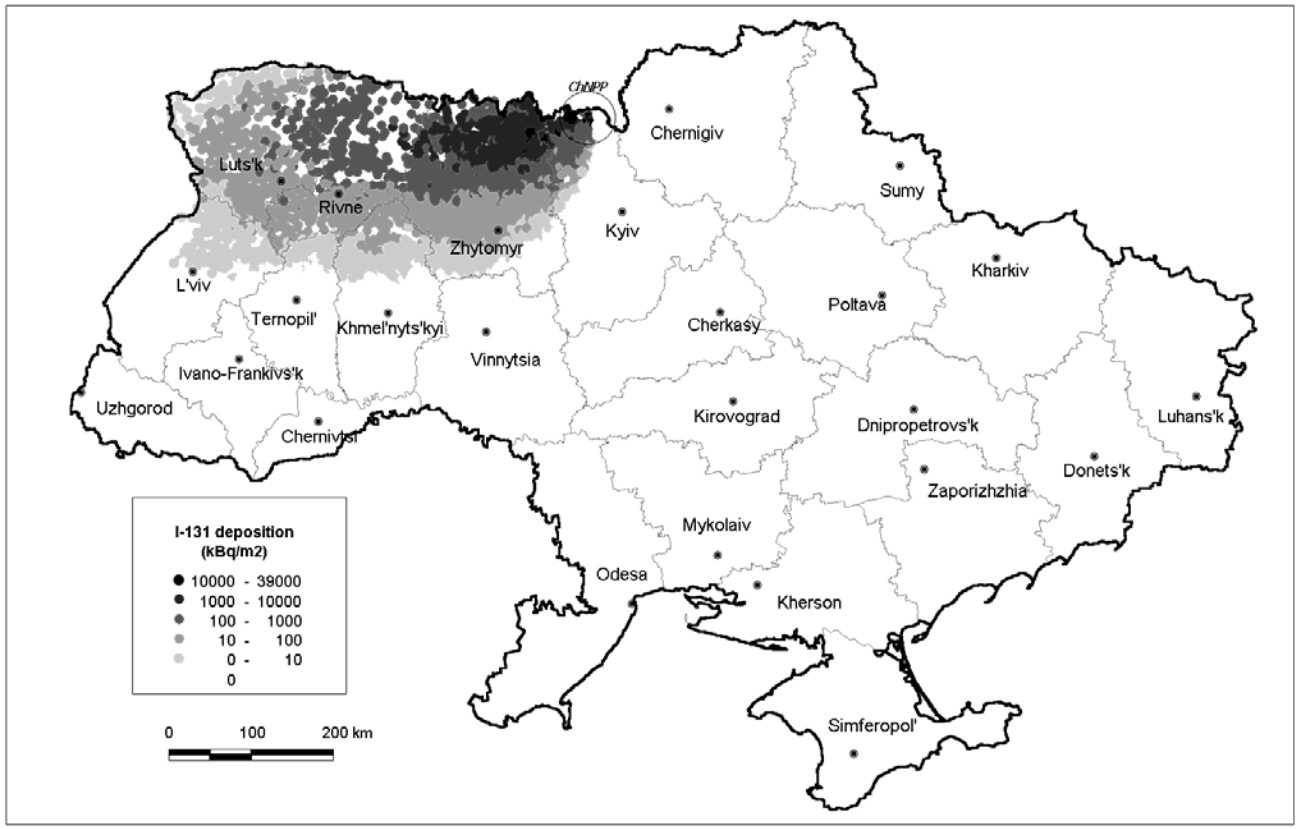

Figure 3.1. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident during 26 April. Separate dot shows deposition value for each settlement.

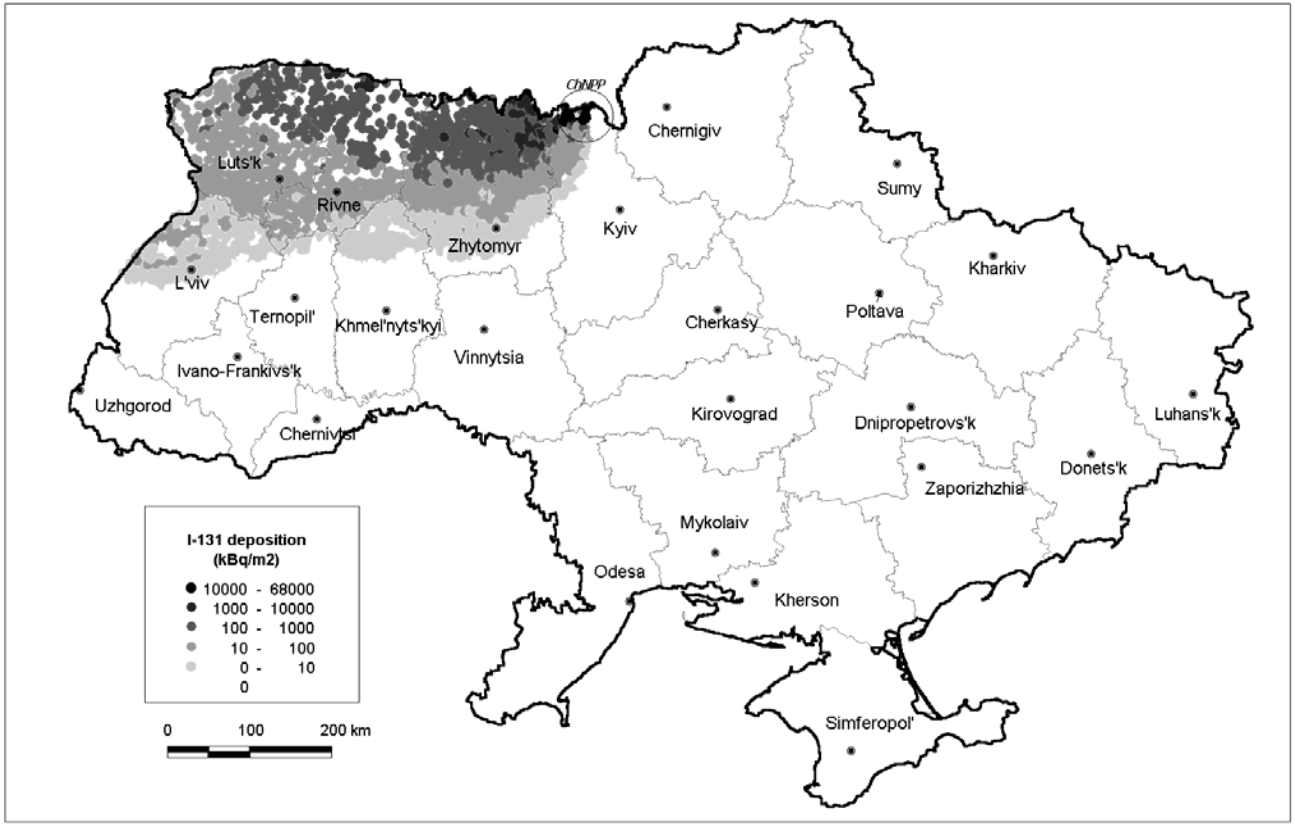

Figure 3.2. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident during 27 April.

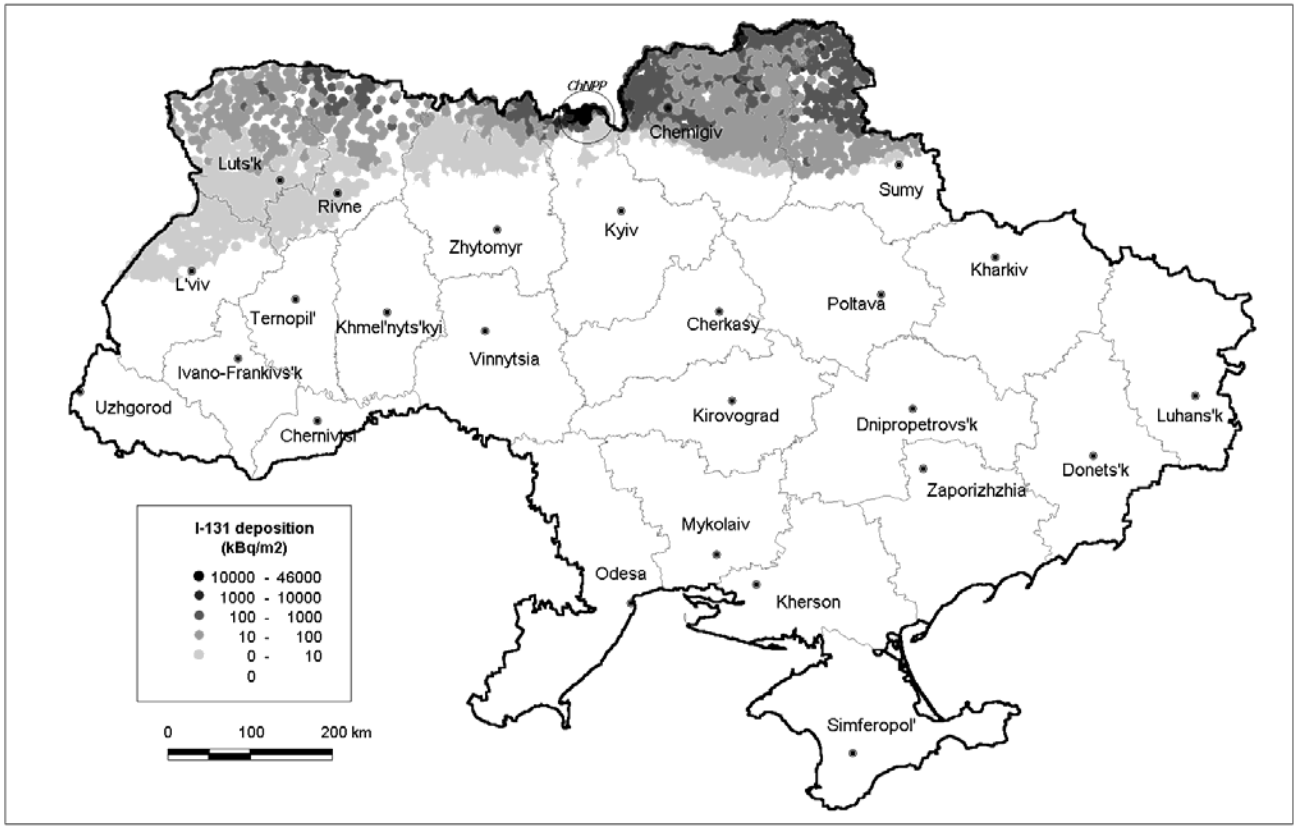

Figure 3.3. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident during 28 April.

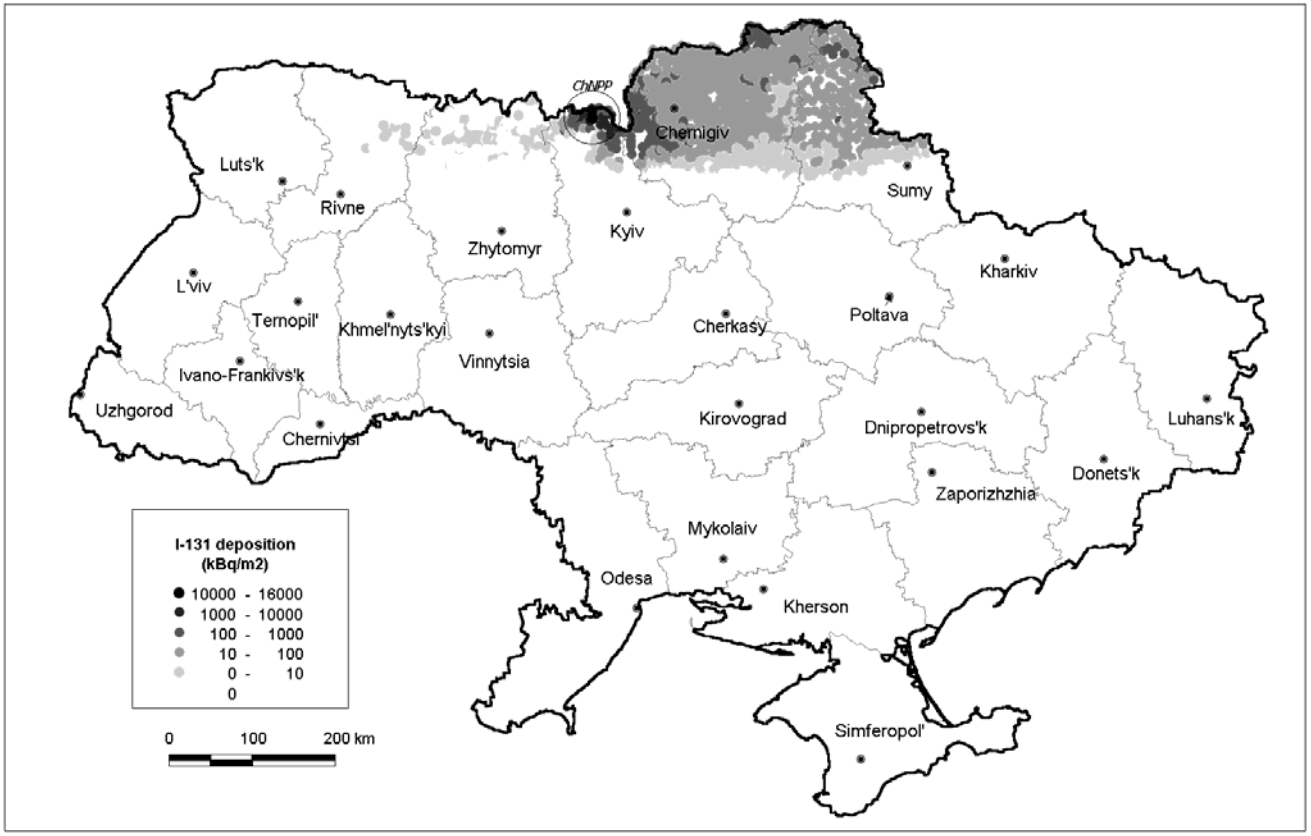

Figure 3.4. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident during 29 April.

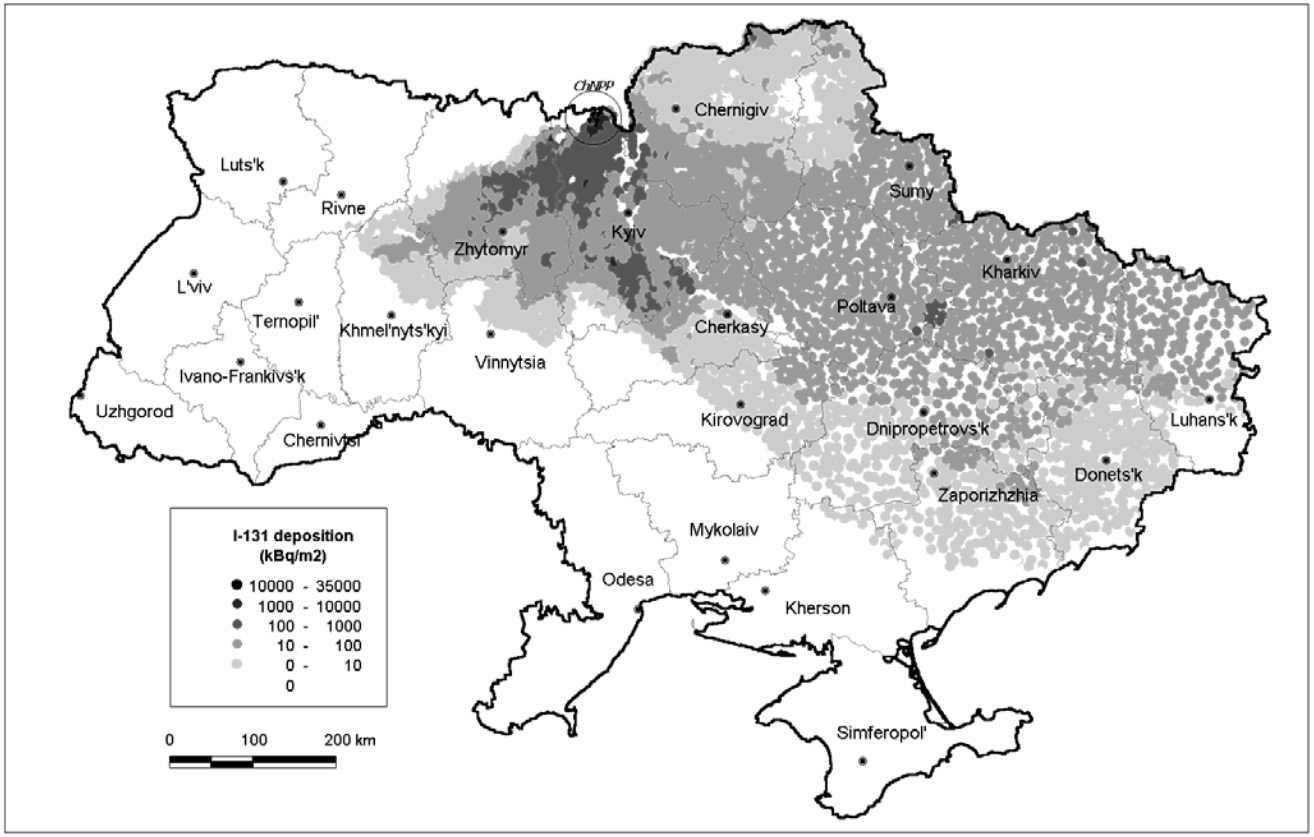

Figure 3.5. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident during 30 April.

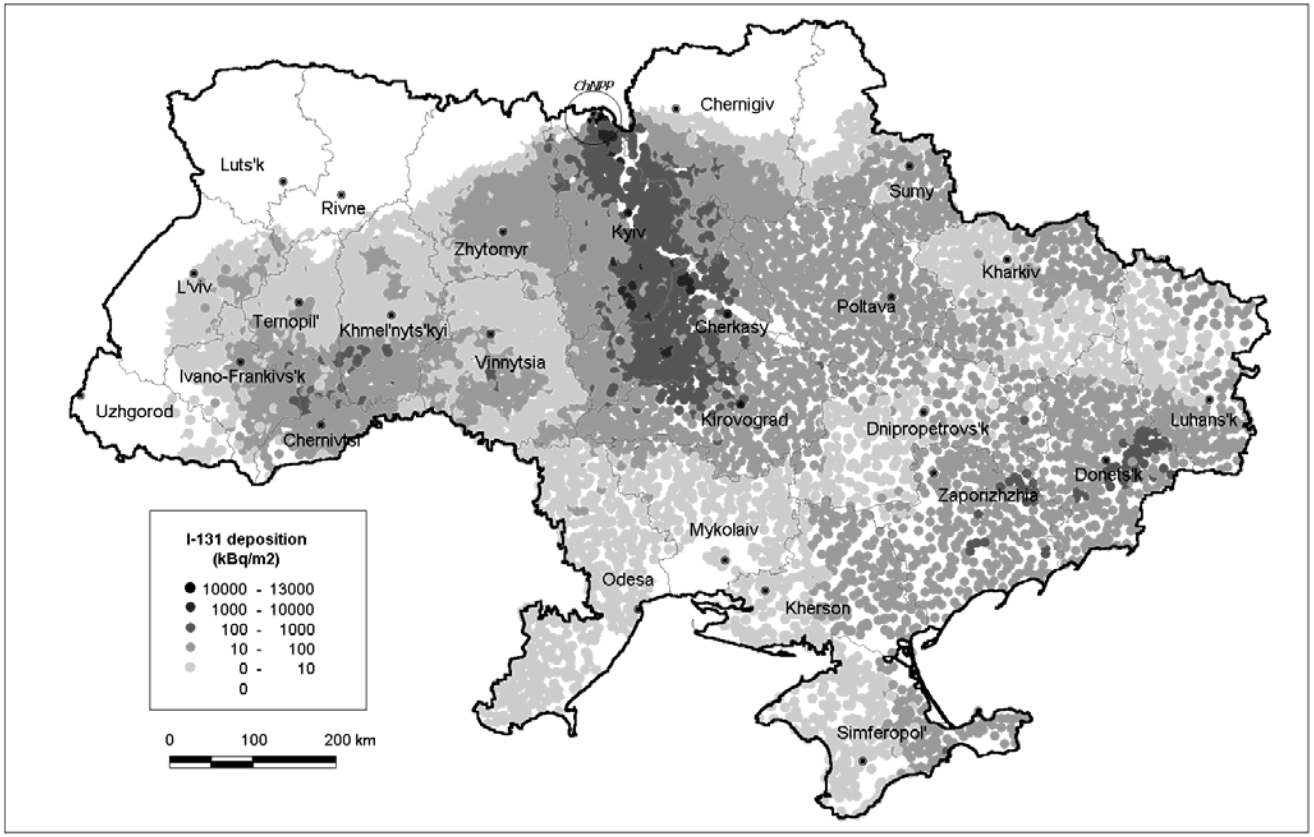

Figure 3.6. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident during 1 May.

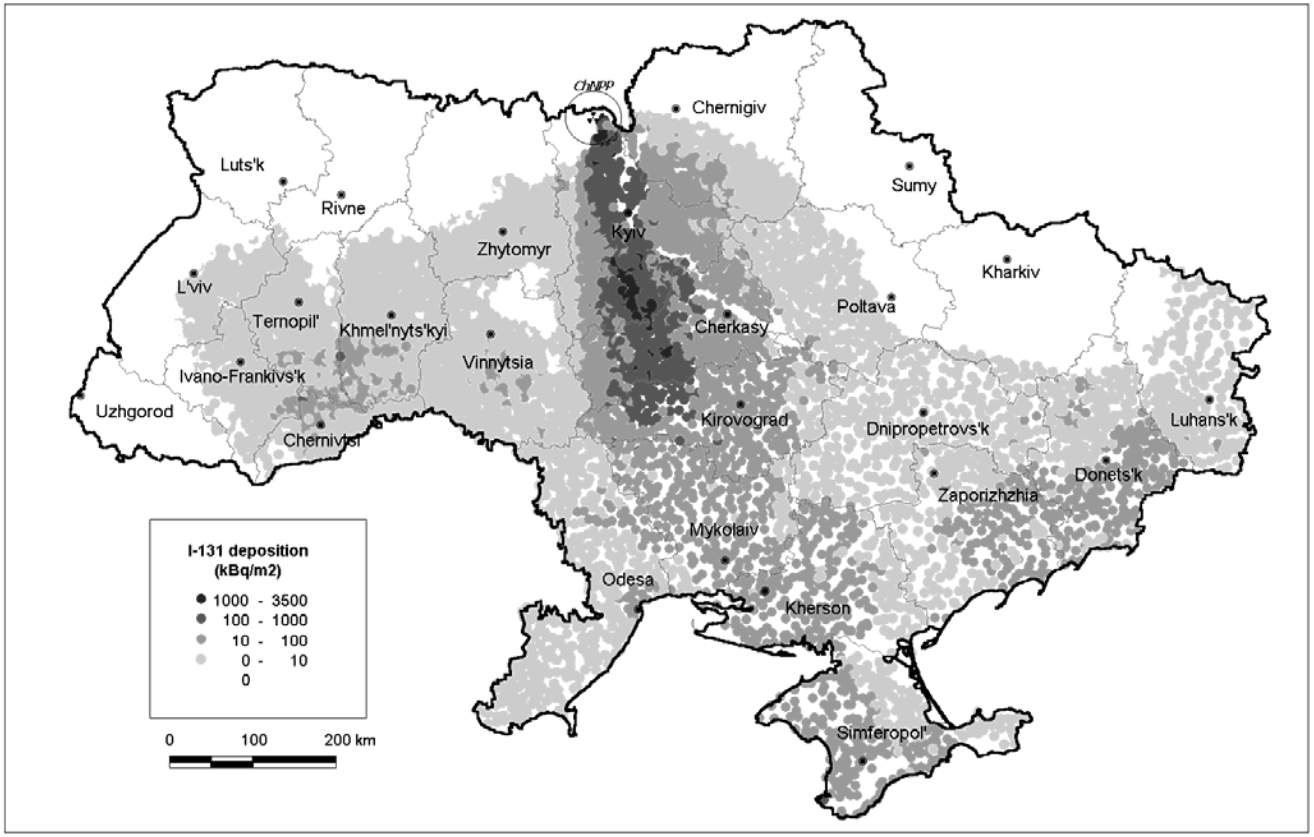

Figure 3.7. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident during 2 May.

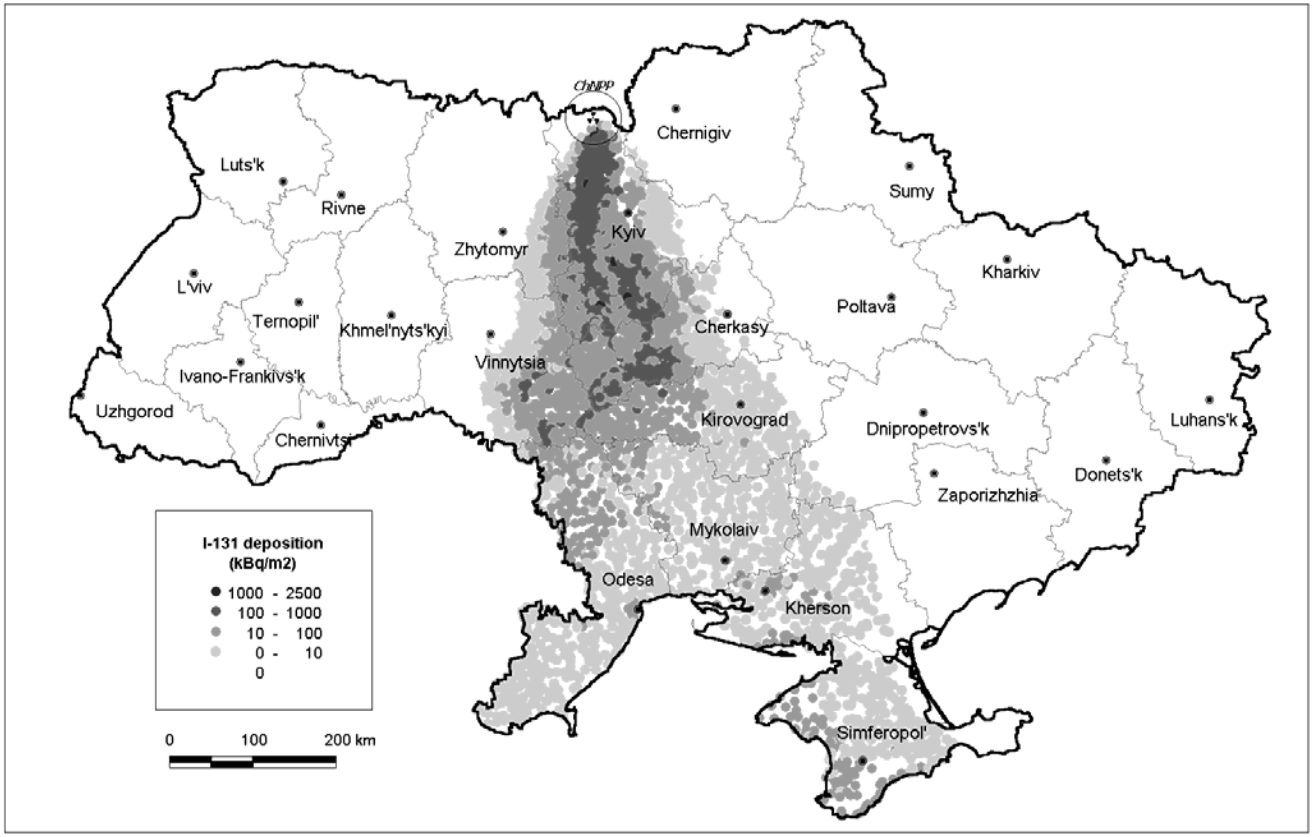

Figure 3.8. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident during 3 May.

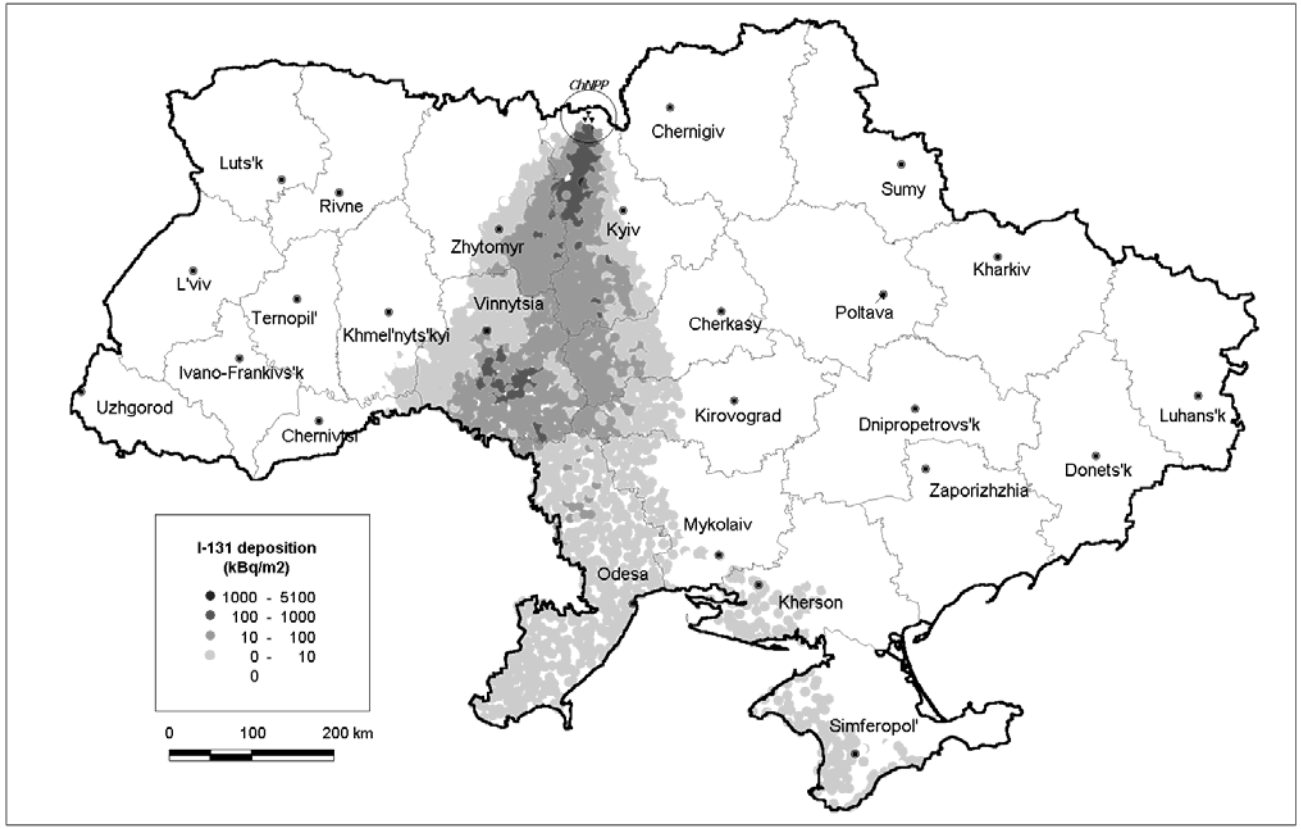

Figure 3.9. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident during 4 May.

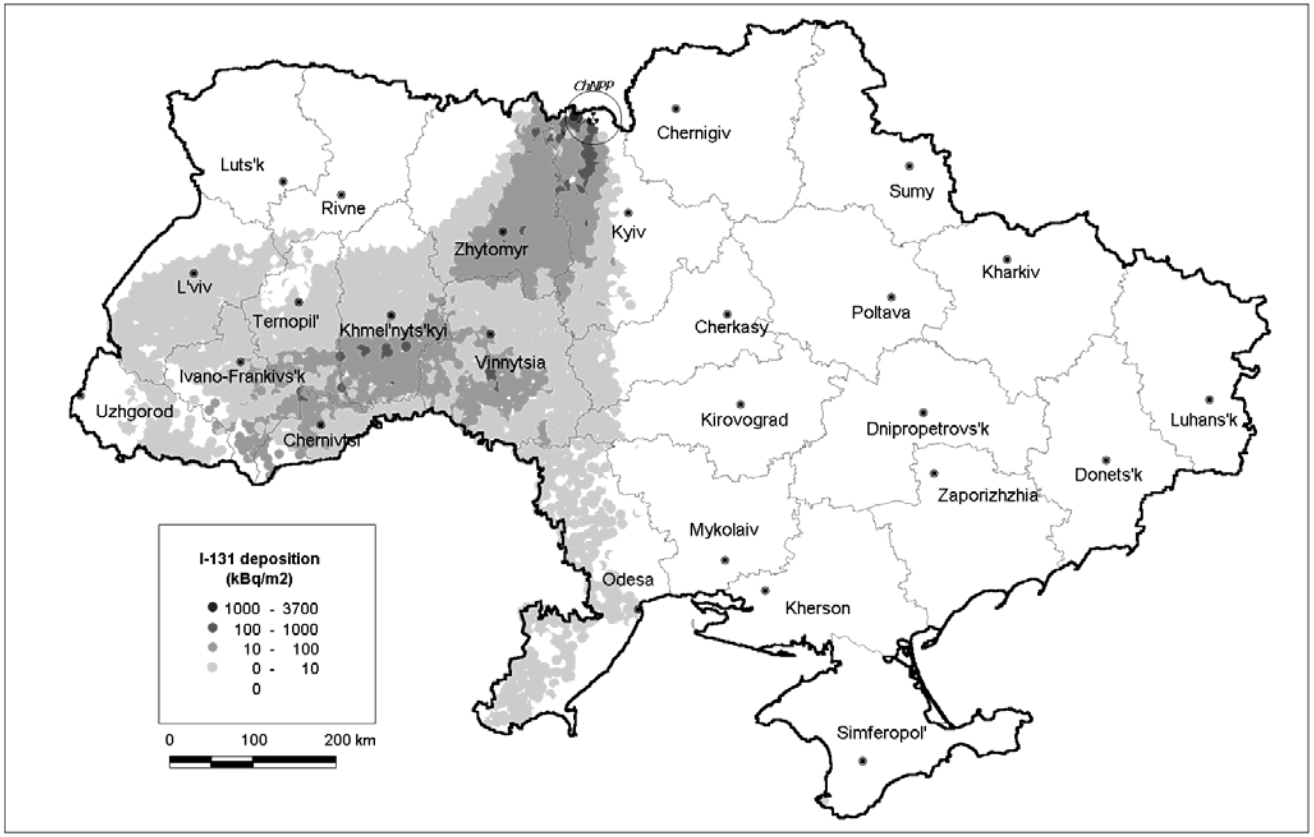

Figure 3.10. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident during 5 May.

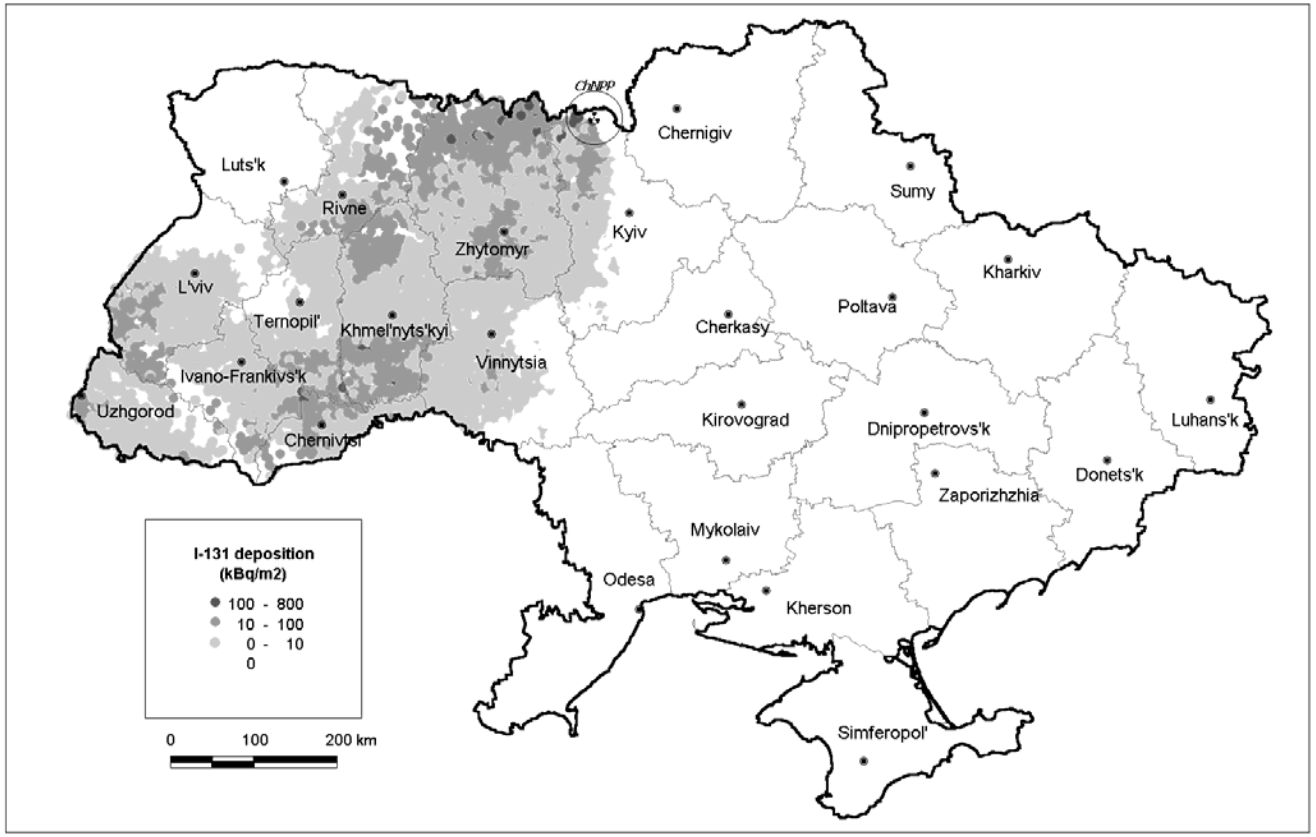

Figure 3.11. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident during 6 May.

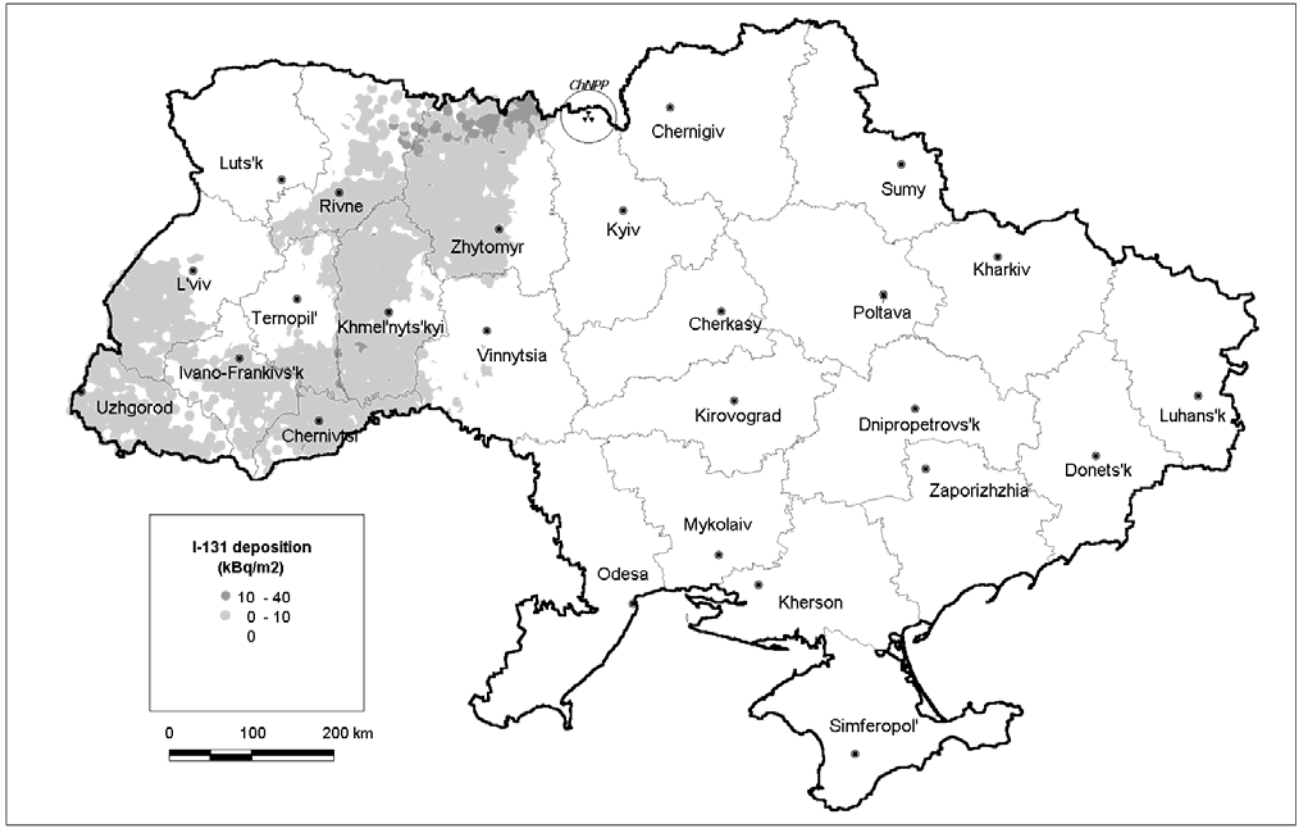

Figure 3.12. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident during 7 May.

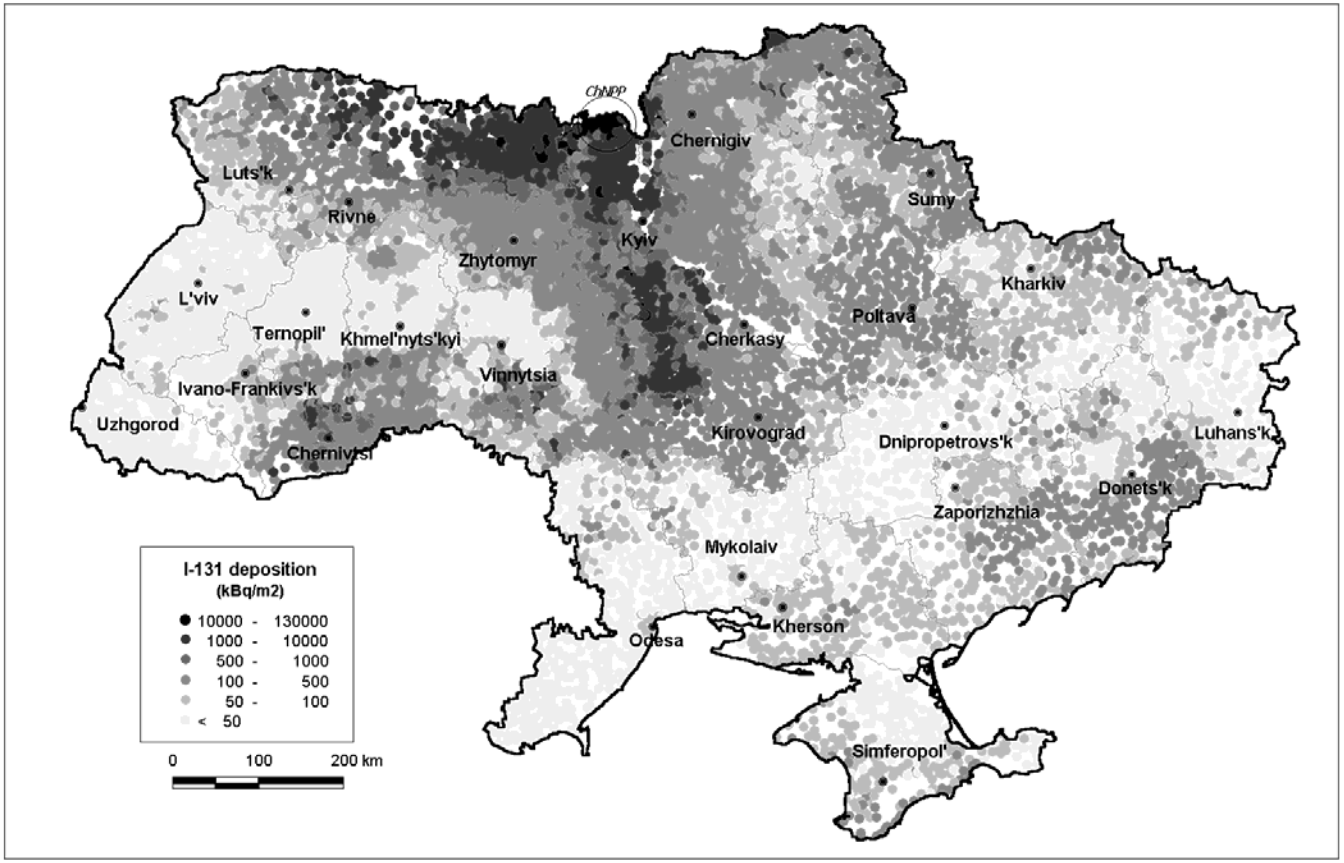

Figure 3.13. Soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident corrected to 26 April. Separate dot shows deposition value for each settlement.

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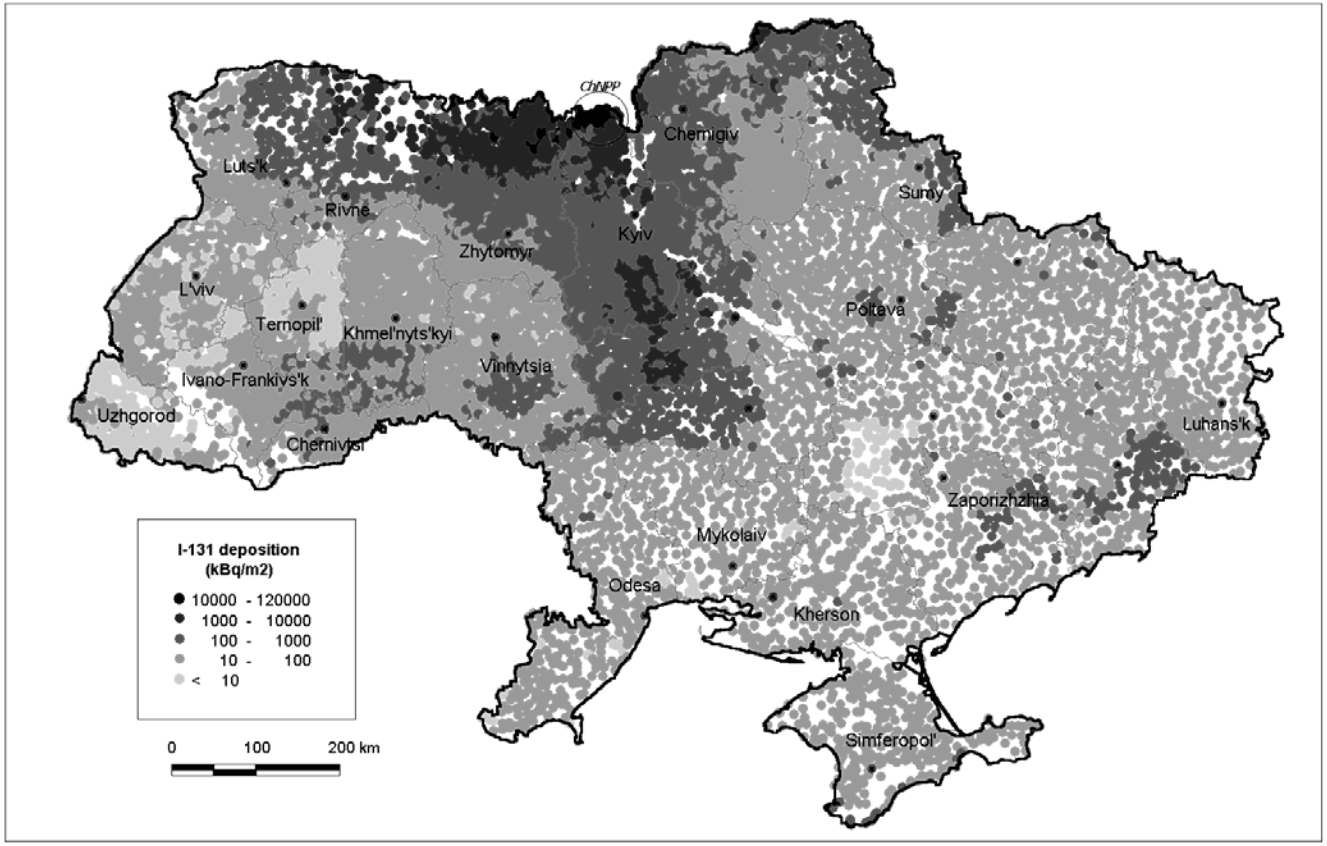

Figure 3.14. Integral soil deposition of ^{131}I in Ukraine (kBq m^{-2}) due to the Chernobyl accident.

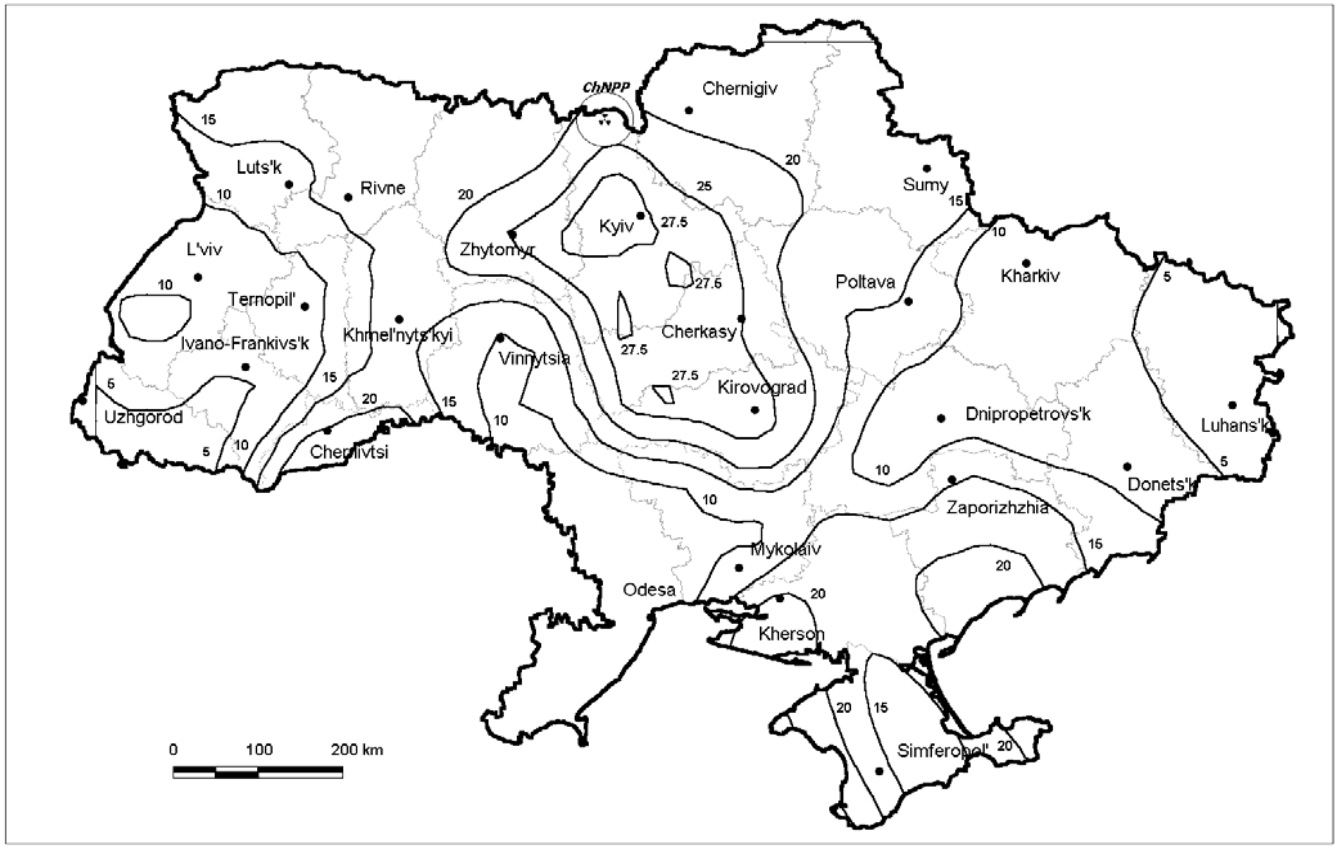

Figure 3.15. Ratio of ^{131}I to ^{137}Cs deposition densities in Ukraine corrected to 26 April 1986.

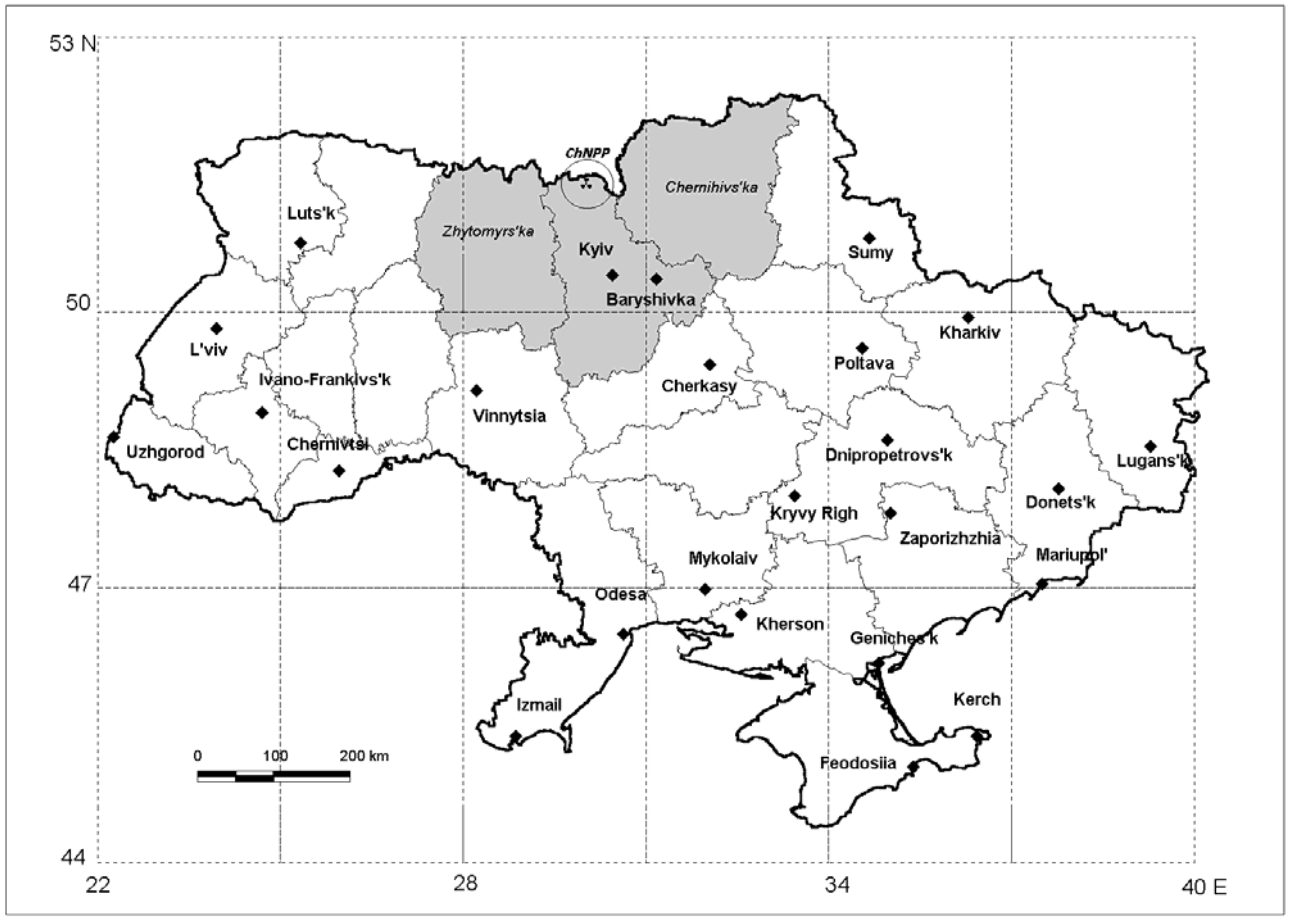

Figure 3.16. ^{131}I daily deposition measurement sites in the territory of Ukraine.

The ^{131}I deposition density field in Ukraine obtained with the help of atmospheric transport modeling of radioactive release from the accidental unit of the Chernobyl nuclear power plant is shown in Figure 3.13. The values of iodine deposition is corrected to the initial day of the accident 26 April 1986. The cumulative ^{131}I deposition density values (a simple sum of daily deposition values) obtained under modeling for settlements in Ukraine are presented in Figure 3.14.

As stated in chapter 2, the deposition field in Ukraine is formed due to dry deposition mainly. The contribution of wet deposition was negligibly low there (with the exception of several small local spots in different regions of Ukraine). According to the simulation results the formation of large-scale deposition spots in the southern part of Kyiv region (near Bila Tserkov) and in Cherkasy region (spot near Cherkasy and Kaniv towns) could be explained the transport and deposition of the activity released from the high-elevated prolonged source taking into account of diurnal variability of turbulent parameters of atmospheric planetary layer. The transport of activity released during night-time of 1 May under conditions of large (more than 10 m s^{-1}) wind velocity resulted in the formation of these spots of radioactive deposition at large (up 200-400 km from the source) distances in this territory.

The largest calculated values of integral ^{131}I deposition (corrected to 26 April 1986) are 127000 kBq m^{-2} in village Varovichi (40 km west from the Chernobyl NPP) and 126000 kBq m^{-2} in village Tolsty Les (22 km west from the Chernobyl NPP). The integral ^{131}I deposition in town Pripjat (2 km north-west from the Chernobyl NPP) was assessed to be 102000 kBq m^{-2}. It's necessary to note that for assessment of deposition in Pripjat a special task of local scale atmospheric transport modeling has been solved. The values of cumulative ^{131}I deposition (a sum of daily depositions without decay correction) in these inhabitant places are 114000 kBq m^{-2}, 115000 kBq m^{-2} and 88200 kBq m^{-2} accordingly. All these inhabitant places are within the exclusion zone of the Chernobyl NPP. The largest values of integral ^{131}I deposition outside the exclusion zone are assessed to be 54600 kBq m^{-2} in Yasen (Kyiv region, 52 km west from the power plant) and 53100 kBq m^{-2} in Martynovichi (Kyiv region, 65 km west). The maximal value of integral ^{131}I deposition in Zhytomyr region is assessed for the village Nove Sharno (65 km from the ChNPP) and equals to 28600 kBq m^{-2}.

In Kyiv the maximal value of integral ^{131}I deposition is 737 kBq m^{-2} and cumulative ^{131}I one is 471 kBq m^{-2}. According modeling in Kyiv the maximal daily averaged air volume ^{131}I concentration was 355 Bq m^{-3} in 1 May, and the air volume concentration averaged over entire period of intensive fallout in Kyiv (from 30 April till 5 May) was 130 Bq m^{-3}. The modeling enabled to obtain assessment of air and ground radioiodine concentration in the high contaminated

territories where there weren't direct measurements made in the initial period of the accident – in Zhytomyr and Chernigiv regions. So, in Zhytomyr town cumulative and integral ^{131}I deposition values are 54 kBq m^{-2} and 91 kBq m^{-2}, in Chernigiv town are 259 kBq m^{-2} and 311 kBq m^{-2}.

The map of ratio of integral (corrected to 26 April 1986) ^{131}I to ^{137}Cs deposition densities in Ukraine as the result of iodine and cesium atmospheric transport and deposition calculations is shown in Figure 3.15. The largest values of this ratio (about 28) is obtained for south trace of radioactive contamination field –in Kyiv, Cherkasy and Kirovograd region. On the whole the ratio decreases as a function of distances from the Chernobyl NPP. The minimal values of ratio is less 5 in Lugans'k region on the east of Ukraine.

The total value of ^{131}I deposition over the territory of Ukraine during the first 12 days after the accident is assessed as 185 PBq. It means 19% of our assessment of total ^{131}I release 975 PBq (estimation of release during the course of the accident) or about 11% of the release estimation made in UNSCARE (2000) or about 6% of the ^{131}I inventory (UNSCARE, 2000).

The main sources of uncertainties of obtained air concentration and deposition density values are variances of activity daily releases from the Chernobyl reactor and dry deposition velocity used under modeling. The another possible source of additional errors under atmospheric transport modeling is meteorological data. The main problem here is the interpolation of wind and temperature measurements made at relatively sparse network into small scale computation grid. It may results in disregarding of the impact of small scale atmospheric processes onto local radioactive air and ground contamination. Another large problem is that the different set of measurement data may contradict one another. For example, according the database of radioactive contamination of soil in territory of Ukraine created on the behalf of the governmental bodies of the country the value of ^{137}Cs soil contamination in Vinnytsia is 20.0 kBq m^{-2} but the measurements of daily ^{137}Cs deposition density made at meteorological station at Vinnytsia gives the value of total cesium deposition as 1.14 kBq m^{-2}, i.e. in 17.5 times lower. The correct and full assessment of uncertainties of obtained results is sufficiently complicated task in this case. So we used the values of differences between the calculated and measured ^{137}Cs deposition values for different inhabitant places (Talerko, 2004) as a main source for the estimation of calculation uncertainties not only for cesium but for another radionuclides including iodine. It enables to take into account the general impact of all sources of possible errors on the local variability of calculated values for every region of Ukraine. As a result the geometric standard deviation (GSD) of a calculated iodine deposition values was estimated as 1.6 for the most regions of Ukraine excluding Mykolaiv, Odesa, Hmel'nitskiy, Chernivtsi, Chernigiv regions (GSD=1.7), Volyn, Ivano-Frankivsk, Ternopil' regions

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(GSD=1.8), Sumy region (GSD=1.9), Lviv and Zakarpatska regions (GSD=2.0).

As stated above, there are few direct measurements of ^{131}I contamination in air and soil were made in Ukraine in the first days after the Chernobyl accident which could be used for the verification of modeling results. On the another hand, there is a quite large set of both direct measurement data and the reconstruction results of ^{131}I contamination for the territory of Belarus. It seems to be useful to apply this approach to simulate the activity transport over the territory of Belarus during the initial period of the accident. It may to obtain more accurate values of iodine release and the picture of ^{131}I deposition field formation for both Belarus and the north-west Ukraine.

3.5. Comparison the modeling results with measurements of ^{129}I soil contamination

The possibility to use the results of direct measurements of long-lived ^{129}I for the reconstruction of ^{131}I deposition pattern have been discussed in Straume *et al*.(1996). This approach have been used by Mironov *et al*. (2002) for reconstruction of the ^{131}I deposition in Belarus and by Pietrzak-Flis Z. *et al*. (2003) for reconstruction of the ^{131}I deposition in Poland after the Chernobyl accident. In last paper the authors determined the thyroid doses for children and adults in Poland using the obtained assessment of ^{131}I deposition. For the territory of Ukraine the measurements of ^{129}I soil contamination have been made by the group from the Center for Radiation Protection and Radioecology of the University Hannover (Germany) and the State University of Agriculture and Ecology (Zhytomyr, Ukraine) (Michel *et al*., 2004). They investigated 42 soil samples taken from high contaminated territory of Zhytomyr region – in 4 villages in Korosten district (120 km south-west of the Chernobyl nuclear power plant) and 3 villages in Narodichi district (about 70 km south-west from the ChNPP). According to the Ukrainian laws these sampling sites in Narodichi district belongs to 2nd contamination zone (including territory with ^{137}Cs deposition densities in the range between 555 kBq m^{-2} and 1480 kBq m^{-2}), and taken sampling sites in Korosten district belongs to 3rd contamination zone (including territory with ^{137}Cs deposition densities in the range between 185 kBq m^{-2} and 555 kBq m^{-2}).

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Table 3.3. ^{137}Cs and ^{131}I deposition densities in inhabitant places of Korosten and Narodichi district (Zhytomyr region, Ukraine) according to atmospheric transport modeling and obtained by Michel (2003) with using ^{129}I measurements. ^{131}I data are corrected to 26 April, 1986.

Village	^{137}Cs deposition (kBq/m^2)						*Cs deposition ratio*	^{131}I deposition (kBq/m^2)		*I deposition ratio*
	Modelling	From database	From Michel (2004)							
			Average for village	Min	Max	Measurements number		Modelling	From Michel (2004)	
Nemirovka	475	506	447	394	498	3	*1.13*	8460	6410	*1.32*
Voronevo	499	539	516	296	851	7	*1.04*	9010	6500	*1.39*
Kupech	383	242	305	277	332	2	*0.79*	4060	6250	*0.65*
Chigiri	512	556	400	237	556	12	*1.39*	9230	4110	*2.25*
Average (Korosten district)	467	461	417				1.10	7690	5818	1.32
Nozdrishche	1320	1098	3908	2587	5229	2	*0.28*	19000	55700	*0.34*
Nove Sharno	1288	1649	3571	3318	3916	3	*0.46*	28600	50500	*0.57*
Hristinovka	1251	911	2478	738	4218	2	*0.37*	15800	43300	*0.36*
Average (Narodichi district)	1287	1219	3319				0.37	21133	49833	0.42

In Table 3.3 the results of ^{131}I atmospheric transport modeling for these inhabitant places are shown compared with the data of ^{131}I contamination reconstruction using ^{129}I measurements from Michel (2004). Besides it, the data of their measurements of ^{137}Cs soil contamination are shown (including measurements number, minimal, maximal and average values for each of 7 villages from Michel (2004) just as the ^{137}Cs deposition values from the database mentioned above. For Korosten district both the values of ^{137}Cs deposition in each village and the value averaged over all 4 inhabitant places are agree quite well. There is a quite good agreement of ^{131}I reconstruction results for 3 of 4 villages (except Chigiri) just as for values averaged over all 4 inhabitant places under the consideration. Meanwhile there is a great discrepancy between ^{137}Cs deposition values obtained by Michel (2004) and taken from the mentioned database (from 2 to 4 times) for each of 3 villages of Narodichi district. The reason of such differences of the ^{137}Cs deposition data is not clear. The data of ^{137}Cs contamination of the database are enough reliable since they were obtained in consequences of large number of measurements in each inhabitant place. The possible explanation is that some samples with the largest values of ^{137}Cs contamination values have been chosen for next analysis in this work. As the result the obtained ^{131}I deposition values in 3 villages of

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Narodichi district are in 2-3 times larger then ones reconstructed using atmospheric transport modeling.

3.6. Comparison the modeling results with measurements of ^{131}I daily deposition measurements

We have compared the calculated values of ^{131}I deposition with results of daily measurements of radioiodine deposition on collectors exposed at meteorological stations of the former USSR in April-May 1986 (Makhon'ko *et al*., 1996). The measurements data at 25 stations located in the territory of Ukraine during the period from 26 April to 7 May 1986 have been used (Figure 3.16). Unfortunately, the closest measurement sites from the Chernobyl nuclear power plant are situated at the distance about 130 km (Kyiv and Baryshevka of Kyiv region). In the territory of another two most contaminated regions of Ukraine (Zhytomyr and Chernigiv ones) there were no sites where the ^{131}I deposition have been measured.

In Figure 3.17 the results of deposition measurements during first 10 days after the accident at some meteorological stations in Ukraine are shown. The results of reconstruction of daily deposition dynamics in these towns on the base of atmospheric transport modeling are given also. On the whole there is a quite good agreement between measurement and modeling data though there are considerable differences at some sites. The values of cumulative ^{131}I deposition during the period of intensive releases from the accidental unit are differs not more then in 1.5 times for 15 meteorological stations, not more then in 3 times - for 19 stations. For the rest 6 stations this ratio is larger then 3 times.

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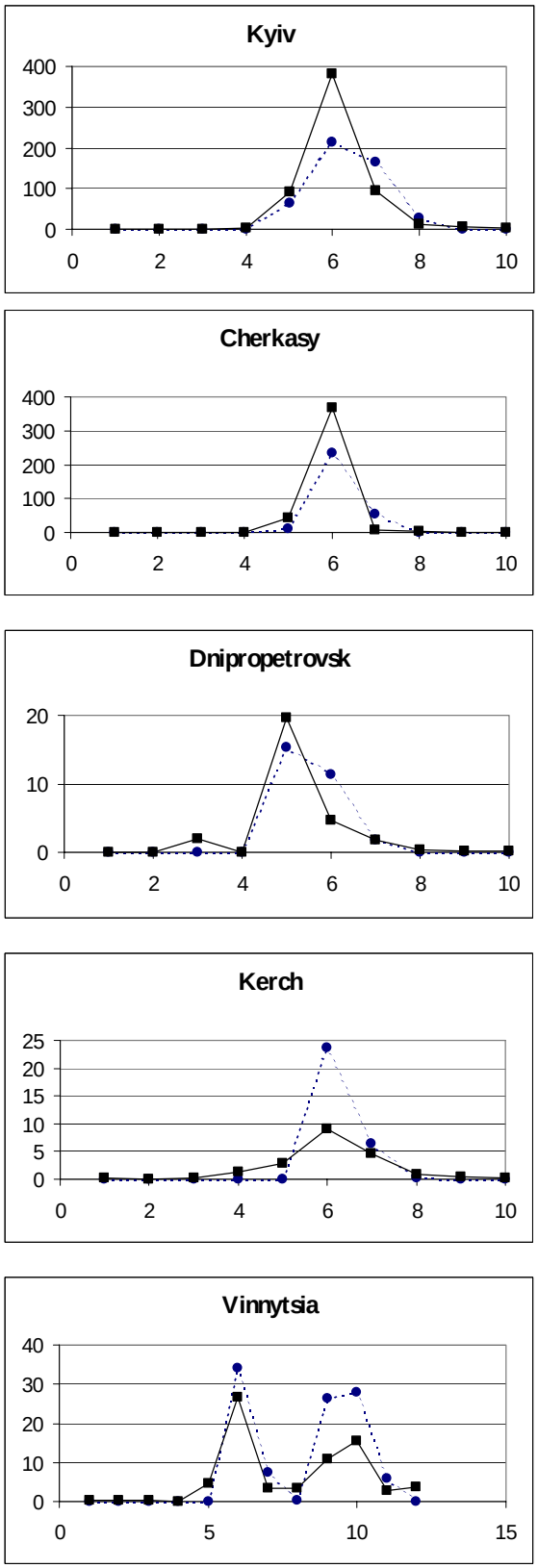

Figure 3.17. The ^{131}I daily depositions measurements (squares, bold lines) and modeling results (circles, dashed lines) (in kBq m^{-2}) as a function of time (in days) for some meteorological stations in Ukraine (1st day – 26 April, 10th – 5 May 1986).

The largest values of ^{131}I daily deposition was obtained for measurement sites within southern part of the radioactive contamination field in Ukraine – Kyiv, Baryshevka and Cherkasy. The dynamics of calculated and measured deposition is similar (the maximal deposition was 30 April - 1 May 1986) for these sites. The ^{131}I cumulative deposition values during first 10 days after the accident are 471 kBq m^{-2} (modeling) and 595 kBq m^{-2} (measurement) in Kyiv, 158 kBq m^{-2} (modeling) and 488 kBq m^{-2} (measurement) in Baryshevka, 301 kBq m^{-2} (modeling) and 425 kBq m^{-2} (measurement) in Cherkasy. There is quite good agreement between calculated and measured radioiodine deposition at meteorological stations in eastern Ukraine – Dnipropetrovsk, Lugans'k, Kerch (Figure 3.17). The deposition values aren't exceed 20-25 kBq m^{-2} per day there. The maximum of air radioactive concentration was on 30 April – 1 May due to arrival of air masses which have been contaminated with releases from the accidental reactor during 28-29 April. This activity moved to northeast, at first, resulting in contamination of the territory of Russia and then turned southward to the territory of eastern Ukraine (chapter 2).

The western part of radioactive contamination field was caused the release during the first day of release 26 April. There is only one measurement site in Lutsk on this territory. The measured value of ^{131}I cumulative deposition is 67 kBq m^{-2} and the modeling result is 83 kBq m^{-2}. But modeling gives 26 April as date of the deposition maximum whereas according measurements the maximal deposition was later - on 28 and 29 April (Figure 3.17). The reason of such differences can be explained by the features of local meteorological conditions which implied on the radioactivity transport and deposition in this region. According both measurements and modeling the period of intensive fallout in south-western part of Ukraine (Vinnytsia, Ivano-Frankivsk) lasted till 7 May and has two maxima – on 1 May and on 5 May.

The values of ^{131}I cumulative deposition calculated for all 25 measurement sites on the territory of Ukraine differ from measured ones not more then in 1.5 times for 15 sites, within interval from 1.5 to 3 times for 6 sites, and for 4 sites these values differs in more then 3 times.

3.7. Conclusions

It was shown that the method of mathematical modeling of radioactivity atmospheric transport and deposition on the underlying surface can be useful tool for the reconstruction of the dynamics of air and ground contamination in the initial period of the Chernobyl accident. This method enables to take into account the features of radioactivity fallout formation including time and space variability of the deposition field.

The map of ^{131}I deposition in the territory of Ukraine caused by the Chernobyl accident was

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obtained using the method of mathematical modeling of radioactive release transport in the atmosphere.

The daily dynamics of radioiodine deposition and air volume concentration is calculated for 12715 inhabited places in Ukraine resulting in the database as a ground for the reconstruction of thyroid doses received by the population of Ukraine. The cumulative ^{131}I deposition values in Kyiv is assessed to be 471 kBq m^{-2}, whereas ^{131}I deposition values in centers of another two the most contaminated regions are 259 kBq m^{-2} in Chernigiv town and 54 kBq m^{-2} in Zhytomyr town. The cumulative ^{131}I deposition in Korosten town (the district center of Zhytomyr region) is assessed to be 4669 kBq m^{-2}.

The used method may be useful for the reconstruction of air and ground deposition concentrations of another volatile nuclides especially short-lived radionuclides for which few measurements data available and which could contribute significantly to thyroid doses of Ukrainian population.

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